

TRANSMITTAL LETTER TO THE UNITED STATES  
DESIGNATED/ELECTED OFFICE (DO/EO/US)  
CONCERNING A FILING UNDER 35 U.S.C. 371

CRZ-96631

U.S. APPLICATION NO. (IF KNOWN, SEE 37 CFR 1.5)

INTERNATIONAL APPLICATION NO.  
PCT/DE96/02658INTERNATIONAL FILING DATE  
19 June 1996PRIORITY DATE CLAIMED  
19 June 1996

## TITLE OF INVENTION

MULTICOMPONENT SYSTEM FOR USE WITH DETERGENT SUBSTANCES

## APPLICANT(S) FOR DO/EO/US

Call, Hans-Peter

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. ☒ This is an express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1).
4. ☐ A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.
5. ☒ A copy of the International Application as filed (35 U.S.C. 371 (c) (2))
  - a. ☒ is transmitted herewith (required only if not transmitted by the International Bureau).
  - b. ☒ has been transmitted by the International Bureau.
  - c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
6. ☐ A translation of the International Application into English (35 U.S.C. 371(c)(2)).
7. ☒ A copy of the International Search Report (PCT/ISA/210).
8. ☐ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371 (c)(3))
  - a. ☐ are transmitted herewith (required only if not transmitted by the International Bureau).
  - b. ☐ have been transmitted by the International Bureau.
  - c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
  - d. ☐ have not been made and will not be made.
9. ☐ A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3))
10. ☐ An oath or declaration of the inventor(s) (35 U.S.C. 371 (c)(4)).
11. ☐ A copy of the International Preliminary Examination Report (PCT/IPEA/409)
12. ☐ A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371 (c)(5)).

## Items 13 to 18 below concern document(s) or information included:

13. ☐ An Information Disclosure Statement under 37 CFR 1.97 and 1.98
14. ☐ An assignment document for recording A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
15. ☐ A **FIRST** preliminary amendment.  
A **SECOND** or **SUBSEQUENT** preliminary amendment.
16. ☐ A substitute specification.
17. ☐ A change of power of attorney and/or address letter.
18. ☒ Certificate of Mailing by Express Mail
19. ☒ Other items or information:

PCT Publication No. WO97/48786

U.S. APPLICATION NO. (IF KNOWN, SEE 37 CFR 1.5)

INTERNATIONAL APPLICATION NO.

ATTORNEY'S DOCKET NUMBER

PCT/DE96/02658

CRZ-96631

20. The following fees are submitted:

CALCULATIONS PTO USE ONLY

**BASIC NATIONAL FEE ( 37 CFR 1.492 (a) (1) - (5)) :**

- ☒ Search Report has been prepared by the EPO or JPO ..... **\$930.00**
- ☐ International preliminary examination fee paid to USPTO (37 CFR 1.482) ..... **\$720.00**
- ☐ No international preliminary examination fee paid to USPTO (37 CFR 1.482) but international search fee paid to USPTO (37 CFR 1.445(a)(2)) ..... **\$790.00**
- ☐ Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO ..... **\$1,070.00**
- ☐ International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfied provisions of PCT Article 33(2)-(4) ..... **\$98.00**

**ENTER APPROPRIATE BASIC FEE AMOUNT =****\$930.00**Surcharge of **\$130.00** for furnishing the oath or declaration later than ☐ 20 ☐ 30 months from the earliest claimed priority date (37 CFR 1.492 (e)).**\$0.00**

CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE
Total claims	- 20 =	0	x \$22.00
Independent claims	- 3 =	0	x \$82.00

**\$0.00****\$0.00**Multiple Dependent Claims (check if applicable). ☐**\$0.00****TOTAL OF ABOVE CALCULATIONS =****\$930.00**Reduction of 1/2 for filing by small entity, if applicable. Verified Small Entity Statement must also be filed (Note 37 CFR 1.9, 1.27, 1.28) (check if applicable). ☐**\$0.00****SUBTOTAL =****\$930.00**Processing fee of **\$130.00** for furnishing the English translation later than ☐ 20 ☐ 30 months from the earliest claimed priority date (37 CFR 1.492 (f)).**\$0.00****TOTAL NATIONAL FEE =****\$930.00**Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31) (check if applicable). ☐**\$0.00****TOTAL FEES ENCLOSED =****\$930.00**Amount to be:  
refunded

\$

charged

\$

☒ A check in the amount of **\$930.00** to cover the above fees is enclosed.

☐ Please charge my Deposit Account No. \_\_\_\_\_ in the amount of \_\_\_\_\_ to cover the above fees.

A duplicate copy of this sheet is enclosed.

☒ The Commissioner is hereby authorized to charge any fees which may be required, or credit any overpayment to Deposit Account No. **18-1215** A duplicate copy of this sheet is enclosed.

**NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.**

SEND ALL CORRESPONDENCE TO:

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SIGNATURE

Karl S. Sawyer, Jr.

NAME

28,902

REGISTRATION NUMBER

February 19, 1998

DATE

**VERIFIED STATEMENT (DECLARATION) CLAIMING SMALL ENTITY  
STATUS (37 CFR 1.9(f) AND 1.27 (b)) - INDEPENDENT INVENTOR**

Docket No.  
CR2-96631

Serial No.

Filing Date

Patent No.

Issue Date

Applicant/ **Hans-Peter Call**  
Patentee:

Invention: **MULTICOMPONENT SYSTEMS FOR USE WITH DETERGENT SUBSTANCES**

As a below named inventor, I hereby declare that I qualify as an independent inventor as defined in 37 CFR 1.9(c) for purposes of paying reduced fees under section 41(a) and (b) of Title 35, United States Code, to the Patent and Trademark Office with regard to the invention entitled above and described in:

- ☐ the specification to be filed herewith.  
☒ the application identified above.  
☐ the patent identified above.

I have not assigned, granted, conveyed or licensed and am under no obligation under contract or law to assign, grant, convey or license, any rights in the invention to any person who could not be classified as an independent inventor under 37 CFR 1.9(c) if that person had made the invention, or to any concern which would not qualify as a small business concern under 37 CFR 1.9(d) or a nonprofit organization under 37 CFR 1.9(e).

Each person, concern or organization to which I have assigned, granted, conveyed, or licensed or am under an obligation under contract or law to assign, grant, convey, or license any rights in the invention is listed below:

- ☒ No such person, concern or organization exists.  
☐ Each such person, concern or organization is listed below.

\*NOTE: Separate verified statements are required from each named person, concern or organization having rights to the invention averring to their status as small entities (37 CFR 1.27)

FULL NAME Dr. Call, Hans-Peter

ADDRESS 59821 Amshery II Kurfuerdenstrasse 6a / Germany  
☒ Individual ☐ Small Business Concern ☐ Nonprofit Organization

FULL NAME

ADDRESS

☐ Individual ☐ Small Business Concern ☐ Nonprofit Organization

FULL NAME

ADDRESS

☐ Individual ☐ Small Business Concern ☐ Nonprofit Organization

FULL NAME

ADDRESS

☐ Individual ☐ Small Business Concern ☐ Nonprofit Organization

000020-70462060

I acknowledge the duty to file, in this application or patent, notification of any change in status resulting in loss of entitlement to small entity status prior to paying, or at the time of paying, the earliest of the issue fee or any maintenance fee due after the date on which status as a small entity is no longer appropriate. (37 CFR 1.28(b))

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application, any patent issuing thereon, or any patent to which this verified statement is directed.

NAME OF INVENTOR Hans-Peter Call

SIGNATURE OF INVENTOR \_\_\_\_\_

DATE: 10.2.98 T.P. Call

NAME OF INVENTOR \_\_\_\_\_

SIGNATURE OF INVENTOR \_\_\_\_\_

DATE: \_\_\_\_\_

NAME OF INVENTOR \_\_\_\_\_

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NAME OF INVENTOR \_\_\_\_\_

SIGNATURE OF INVENTOR \_\_\_\_\_

DATE: \_\_\_\_\_

PTO/PCT Rec'd 08 FEB 2000  
09/029401

Patent

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of

Docket: 21373.001

Hans-Peter CALL

Serial No. 09/029,401

Group Art Unit: To be assigned

Filed: February 19, 1998

Examiner: To be assigned

For: MULTICOMPONENT SYSTEM FOR USE WITH DETERGENT SUBSTANCES

**PRELIMINARY AMENDMENT**

Assistant Commissioner of  
Patents and Trademarks  
Washington, D.C. 20231



Dear Sir:

Preliminary to the examination of the above identified application, please enter the following preliminary amendment.

**IN THE SPECIFICATION:**

On page 1, line 3, please insert --FIELD OF THE INVENTION--.

On page 1, line 5, please insert --BACKGROUND OF THE INVENTION--.

On page 3, line 10, please delete "General Description of the Invention" and insert --BRIEF SUMMARY OF THE INVENTION--.

On page 4, line 14, please insert --DETAILED DESCRIPTION OF THE INVENTION--.

On page 50, line 1, please insert --IN THE ABSTRACT--.

I hereby certify that this correspondence is being deposited with the United States Postal Service on this date, Feb. 4, 2000 in an envelope as "First Class Mail Post Office to Addressee" addressed to the Assistant Commissioner of Patents and Trademarks, Washington, D.C. 20231.

Christy B. Edwards

008020 "T0462050

On page 50, line 3, please insert the following:

-- A multicomponent system for use with detergents comprising at least one suitable oxidizing agent; at least one mediator selected from the group consisting of hydroxylamines, hydroxylamine derivatives, hydroxamic acids, hydroxamic acid derivatives, and aliphatic, cycloaliphatic, heterocyclic or aromatic compounds that contain at least one N-hydroxy, oxime, N-oxy, or N,N'-dioxy function; and at least one comediator selected from the group consisting of aryl-substituted alcohols, carbonyl compounds, aliphatic ethers, phenol ethers, and olefins (alkenes). The multicomponent system may further comprise at least one oxidation catalyst and/or a predetermined amount of at least one free amine of a respective mediator.--

#### IN THE CLAIMS:

Please amend the claims as follows:

1. (Amended) A multicomponent system for use with detergents[, containing] comprising
  - a) optionally, at least one oxidation catalyst;
  - b) at least one suitable oxidizing agent;
  - c) at least one mediator, selected from the group [comprising] consisting of hydroxylamines, hydroxylamine derivatives, hydroxamic acids, hydroxamic acid derivatives, and [the] aliphatic, cycloaliphatic, heterocyclic or aromatic compounds that contain at least one N-hydroxy, oxime, N-oxy, or N,N'-dioxy function; and
  - d) at least one comediator, selected from the group [comprising] consisting of aryl-substituted alcohols, carbonyl compounds, aliphatic ethers, phenol ethers, and[/or] olefins (alkenes)[; and]
  - [e) optionally, a small quantity of at least one free amine of a respective mediator used].

2. (Amended) The multicomponent system of claim 1, [characterized in that in addition to the these substances, it contains] further comprising phenolic and/or nonphenolic compounds with one or more benzene nuclei.

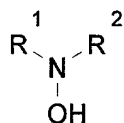
3. (Amended) The multicomponent system of claim 1, wherein the oxidation catalyst is selected from the group consisting of [or 2, characterized in that as the oxidation catalyst, it contains] one or more oxidoreductases of classes 1.1.1 - 1.97.

4. (Amended) The multicomponent system of claim 3, wherein the system comprises [characterized in that it contains] one or more oxidoreductases which use oxygen, peroxides or quinones as electron acceptors.

5. (Amended) The multicomponent system of claim 3, [characterized in that as the oxidoreductase, it contains a laccase (1.10.3.2.)] wherein the oxidoreductase is a laccase of class 1.10.3.2.

6. (Amended) The multicomponent system of claim 1 [or 2], [characterized in that] wherein component c) comprises the aliphatic, cycloaliphatic, heterocyclic or aromatic compounds that contain at least one N-hydroxy, oxime, N-oxy and N,N'-dioxy function [, as the NO-, NOH- or H-NR- OH-containing aliphatic, cycloaliphatic, heterocyclic or aromatic compounds, it contains N-hydroxy, oxime, N-oxy and N,N'-dioxy compounds] in single- or multicomponent systems.

7. (Amended) The multicomponent system of claim 6, wherein the component c), [characterized in that component c)], as the NO-, NOH- or H-NR-OH- containing compounds, [contains] comprises hydroxylamines of the general formula I

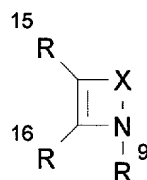


in which the substituents R<sup>1</sup> and R<sup>2</sup>, which may be the same or different, independently of one other represent one of the following groups: hydrogen, C<sub>1</sub>-C<sub>12</sub> alkyl, carbonyl C<sub>1</sub>-C<sub>6</sub> alkyl, phenyl, aryl, of which C<sub>1</sub>-C<sub>12</sub> alkyl, carbonyl C<sub>1</sub>-C<sub>6</sub> alkyl, phenyl, aryl may be unsubstituted or may also be substituted once or multiple times with the radical R<sup>3</sup>,

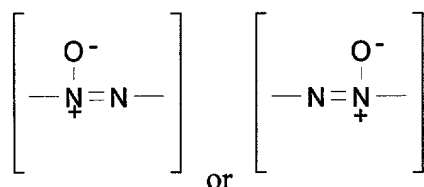
in which the radical R<sup>3</sup> may represent one of the following groups: hydrogen, halogen, hydroxyl, formyl, carboxyl and salts and esters thereof, amino, nitro, C<sub>1</sub>-C<sub>12</sub> alkyl, C<sub>1</sub>-C<sub>6</sub> alkyloxy, carbonyl C<sub>1</sub>-C<sub>6</sub> alkyl, phenyl, sulfono, their esters and salts, sulfamoyl, carbamoyl, phospho, phosphono, phosphonooxy and their salts and esters; in which the amino, carbamoyl and sulfamoyl groups of the radical R<sup>3</sup> may be unsubstituted or may be substituted [once] one or two times with hydroxyl, C<sub>1</sub>-C<sub>3</sub> alkyl, C<sub>1</sub>-C<sub>3</sub> alkoxy; in which the radicals R<sup>1</sup> and R<sup>2</sup> can jointly form a group-B-, and -B- in that case represents one of the following groups: (-CHR<sup>4</sup>-)<sub>n</sub>, (CR<sup>4</sup>=CH-)<sub>m</sub>; and in which R<sup>4</sup> is a substituent that is defined [line] like R<sup>3</sup>, and n represents an integer from 1 to 6 and m represents an integer from 1 to 3.

8. (Amended) The multicomponent system of claim 6, wherein component c) [characterized in that component c)], as the NO-, NOH- or H-NR-OH- containing compounds, [contains] comprises substances of the general formula II





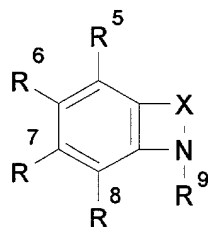
in which X stands for one of the following groups:  $(-N=N-)$ ,  $(-N=CR^{10}-)_p$ ,  $(-CR^{10}=N-)_p$ ,  $(-CR^{11}=CR^{12}-)_p$



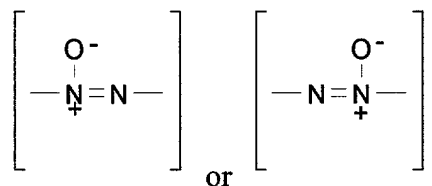
and p is equal to 1 or 2, in which the radicals  $R^9$  to  $R^{12}$ ,  $R^{15}$  and  $R^{16}$  may be the same or different and independently of one another can represent one of the following groups: hydrogen, halogen, hydroxyl, formyl, carboxyl and salts and esters thereof; amino, nitro,  $C_1$ - $C_{12}$  alkyl,  $C_1$ - $C_6$  alkyloxy, carbonyl  $C_1$ - $C_6$  alkyl, phenyl, sulfono esters and salts thereof, sulfamoyl, carbamoyl, phospho, phosphono, phosphonooxy and their salts and esters; and in which the amino, carbamoyl and sulfamoyl groups of the radicals  $R^9$  to  $R^{12}$ ,  $R^{15}$  and  $R^{16}$  may be unsubstituted or may also be substituted [once] one or two times with hydroxyl,  $C_1$ - $C_3$  alkyl,  $C_1$ - $C_3$  alkoxy; and in which the radicals  $R^{15}$  and  $R^{16}$  can form a common group  $-G-$ , and  $-G-$  represents one of the following groups:  $(-CR^5=CR^6-CR^7=CR^8-)$  or  $(-CR^8=CR^7-CR^6=CR^5-)$ , in which the radicals  $R^5$  to  $R^8$  may be the same or different and independently of one another can represent one of the following groups: hydrogen, halogen, hydroxyl, formyl, carboxyl and salts and esters thereof; amino, nitro,  $C_1$ - $C_{12}$  alkyl,  $C_1$ - $C_6$  alkyloxy, carbonyl  $C_1$ - $C_6$  alkyl, phenyl, sulfono, esters and salts thereof, sulfamoyl, carbamoyl, phospho, phosphono, phosphonooxy and their salts and esters, and in which the amino, carbamoyl and sulfamoyl groups of the radicals  $R^5$  to  $R^8$  may be unsubstituted or may also be substituted [once] one or two times with hydroxyl,  $C_1$ - $C_3$  alkyl,  $C_1$ -

C<sub>3</sub> alkoxy; and in which the C<sub>1</sub>-C<sub>12</sub> alkyl, C<sub>1</sub>-C<sub>6</sub> alkyloxy, carbonyl C<sub>1</sub>-C<sub>6</sub> alkyl, phenyl, aryl groups of radicals R<sup>5</sup> to R<sup>8</sup> may be unsubstituted or may also be substituted one or two times with the radical R<sup>18</sup>; in which the radical R<sup>18</sup> can represent one of the following groups: hydrogen, halogen, hydroxyl, formyl, carboxyl and their salts and esters; amino, nitro, C<sub>1</sub>-C<sub>12</sub> alkyl, C<sub>1</sub>-C<sub>6</sub> alkyloxy, carbonyl C<sub>1</sub>-C<sub>6</sub> alkyl, phenyl, aryl, and their esters and salts, and the carbamoyl, sulfamoyl, amino groups of the radical R<sup>18</sup> may be unsubstituted or may also be substituted [once] one or two times with the radical R<sup>19</sup> and the radical R<sup>19</sup> may represent one of the following groups: hydrogen; hydroxyl, formyl, carboxyl and their salts and esters; amino, nitro, C<sub>1</sub>-C<sub>12</sub> alkyl, C<sub>1</sub>-C<sub>6</sub> alkyloxy, carbonyl C<sub>1</sub>-C<sub>6</sub> alkyl, phenyl, aryl.

9. (Amended) The multicomponent system of claim 6, wherein component c) [characterized in that the component c)], as the NO-, NOH- or H-NR- OH-containing compounds, [contains] comprises a compound[s] of the general formula III,



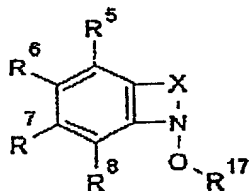
in which X stands for one of the following groups: (-N=N-), (-N=CR<sup>10</sup>-)<sub>p</sub>, (-CR<sup>10</sup>=N-)<sub>p</sub>, (-CR<sup>11</sup>=CR<sup>12</sup>-)<sub>p</sub>



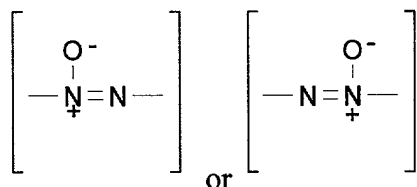
and p is equal to 1 or 2,

in which the radicals  $R^1$  to  $R^{12}$  are same or different and independently of one another can represent one of the following groups: hydrogen, halogen, hydroxyl, formyl, carboxyl and salts and esters thereof; amino, nitro,  $C_1$ - $C_{12}$  alkyl,  $C_1$ - $C_6$  alkyloxy, carbonyl  $C_1$ - $C_6$  alkyl, phenyl, aryl, sulfono, esters and salts thereof, sulfamoyl, carbamoyl, phospho, phosphono, phosphonooxy and their salts and esters; and in which their amino, carbamoyl and sulfamoyl groups may be unsubstituted or may also be substituted [once] one or two times with hydroxy,  $C_1$ - $C_3$  alkyl,  $C_1$ - $C_3$  alkoxy; and in which the  $C_1$ - $C_{12}$  alkyl,  $C_1$ - $C_6$  alkyloxy, carbonyl  $C_1$ - $C_6$  alkyl, phenyl, aryl, aryl  $C_1$ - $C_6$  alkyl groups of radicals  $R^5$  to  $R^{12}$  may be unsubstituted or substituted [once] one or two times with the radical  $R^{13}$ , and the radical  $R^{13}$  can represent one of the following groups: hydrogen, halogen, hydroxyl, formyl, carboxyl and their salts and esters; amino, nitro,  $C_1$ - $C_{12}$  alkyl,  $C_1$ - $C_6$  alkyloxy, carbonyl  $C_1$ - $C_6$  amino, nitro,  $C_1$ - $C_{12}$  alkyl,  $C_1$ - $C_6$  alkyloxy, carbonyl  $C_1$ - $C_6$  alkyl, phenyl, aryl, sulfono, sulfeno, sulfino, and their esters and salts; the [The] carbamoyl, sulfamoyl, amino groups of the radical  $R^{13}$  may be unsubstituted or may also be substituted [once] one or two times with the radical  $R^{14}$  [The] ; the radical  $R^{14}$  may represent one of the following groups: hydrogen; hydroxyl, formyl, carboxyl and their salts and esters; amino, nitro,  $C_1$ - $C_{12}$  alkyl,  $C_1$ - $C_6$  alkyloxy, carbonyl  $C_1$ - $C_6$  alkyl, phenyl or aryl.

10. (Amended) The multicomponent system of claim 6, wherein component c) [characterized in that the component c)], as the NO-, NOH- or H-NR- OH-containing compounds, [contains compounds] , comprises a compound of the general formula IV,



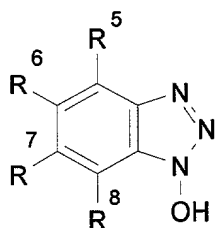
in which X stands for one of the following groups:  $(-N=N-)$ ,  $(-N=CR^{10}-)_p$ ,  $(-CR^{10}=N-)_p$ ,  $(-CR^{11}=CR^{12}-)_p$



and  $p$  is equal to 1 or 2,

in which [for] the radicals R<sup>5</sup> to R<sup>8</sup> and R<sup>10</sup> to R<sup>12</sup> [the same as in claim 9 applies] are defined as above, and R<sup>17</sup> can be hydrogen, C<sub>1</sub>-C<sub>10</sub> alkyl, C<sub>1</sub>- C<sub>10</sub> carbonyl, of which C<sub>1</sub>-C<sub>10</sub> alkyl and C<sub>1</sub>-C<sub>10</sub> carbonyl can be unsubstituted or mono- or polysubstituted with a radical R<sup>18</sup>, which is defined like R<sup>3</sup>.

11. (Amended) The multicomponent system of claim 6, wherein the component c)  
[characterized in that component c)], as the NO-, NOH- or H-NR-OH- containing compounds,  
[contains] comprises 1-hydroxybenzotriazol and [the] tautomeric benzotriazole-1-oxide, [as well  
as] in addition to their esters and salts, of the formula V



in which the radicals R<sup>1</sup> to R<sup>8</sup> may be the same or different and independently of one another can represent one of the following groups: hydrogen, halogen, hydroxyl, formyl, carboxyl and salts

and esters thereof; amino, nitro, C<sub>1</sub>-C<sub>12</sub> alkyl, C<sub>1</sub>-C<sub>6</sub> alkyloxy, carbonyl C<sub>1</sub>-C<sub>6</sub> alkyl, phenyl, sulfono esters and salts thereof, sulfamoyl, carbamoyl, phospho, phosphono, phosphonooxy and their salts and esters; and in which the amino, carbamoyl and sulfamoyl groups of the radicals R<sup>5</sup> to R<sup>8</sup> may be unsubstituted or may also be substituted [once] one or two times with hydroxyl, C<sub>1</sub>-C<sub>3</sub> alkyl, C<sub>1</sub>-C<sub>3</sub> alkoxy; and in which the C<sub>1</sub>-C<sub>12</sub> alkyl, C<sub>1</sub>-C<sub>6</sub> alkyloxy, carbonyl C<sub>1</sub>-C<sub>6</sub> alkyl, phenyl, aryl groups of radicals R<sup>5</sup> to R<sup>8</sup> may be unsubstituted or may also be substituted or mono- or polysubstituted with the radical R<sup>18</sup>, in which the radical R<sup>18</sup> can represent one of the following groups: hydrogen, halogen, hydroxyl, formyl, carboxyl and their salts and esters; amino, nitro, C<sub>1</sub>-C<sub>12</sub> alkyl, C<sub>1</sub>-C<sub>6</sub> alkyloxy, carbonyl C<sub>1</sub>-C<sub>6</sub> alkyl, phenyl, aryl, sulfono, sulfeno, sulfino, and their esters and salts, and the carbamoyl, sulfamoyl, amino groups of the radical R<sup>18</sup> may be unsubstituted or may also be substituted [once] one or two times with the radical R<sup>19</sup>, and the radical R<sup>19</sup> may represent one of the following groups: hydrogen, hydroxyl, formyl, carboxyl and their salts and esters; amino, nitro, C<sub>1</sub>-C<sub>12</sub> alkyl, C<sub>1</sub>-C<sub>6</sub> alkyloxy, carbonyl C<sub>1</sub>-C<sub>6</sub> alkyl, phenyl, aryl.

12. (Amended) The multicomponent system of claim 6, wherein component c) [characterized in that component c)], as the NO-, NOH- or H-NR-OH- containing compounds, [contains such] comprises compounds of azoles.

13. (Amended) The multicomponent system of claim 6, wherein component c) [characterized in that component c)], as the NO-, NOH- or H-NR-OH- containing compounds, [contains such]

comprises compounds of condensed heterocyclic compounds which [contain] comprise a triazolo or tetrazolo unit[, such as:] selected from the group consisting of:

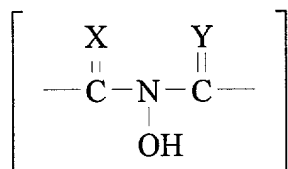
[1,2,4]triazolo[4,3-a]pyridine,  
 [1,2,4]triazolo[1,5-a]pyridine,  
 [1,2,4]triazolo[4,3-a]quinoline,  
 [1,2,4]triazolo[4,3-b]isoquinoline,  
 [1,2,4]triazolo[3,4-a]isoquinoline,  
 [1,2,4]triazolo[1,5-b]isoquinoline,  
 [1,2,4]triazolo[5,1-a]isoquinoline,  
 [1,2,3]triazolo[1,5-a]pyridine,  
 [1,2,3]triazolo[4,5-b]pyridine,  
 [1,2,3]triazolo[4,5-c]pyridine,  
 [1,2,3]triazolo[1,5-a]quinoline,  
 [1,2,3]triazolo[5,1-a]isoquinoline,  
 [1,2,4]triazolo[4,3-b]pyridazine,  
 [1,2,4]triazolo[1,5-b]pyridazine,  
 [1,2,4]triazolo[4,5-d]pyridazine,  
 [1,2,4]triazolo[4,3-b]quinoline,  
 [1,2,4]triazolo[3,4-a]phthalazine,  
 [1,2,4]triazolo[4,3-a]pyrimidine,  
 [1,2,4]triazolo[4,3-c]pyrimidine,  
 [1,2,4]triazolo[1,5-a]pyrimidine,  
 [1,2,4]triazolo[1,5-c]pyrimidine,

[1,2,4]triazolo[4,3-c]quinazoline,  
 [1,2,4]triazolo[1,4-a]quinazoline,  
 [1,2,4]triazolo[1,5-c]quinazoline,  
 [1,2,4]triazolo[5,1-b]quinazoline,  
 [1,2,3]triazolo[1,5-a]pyrimidine,  
 [1,2,3]triazolo[1,5-c]pyrimidine,  
 [1,2,3]triazolo[4,5-d]pyrimidine,  
 [1,2,3]triazolo[1,5-a]quinazoline,  
 [1,2,3]triazolo[1,5-c]quinazoline,  
 [1,2,4]triazolo[4,3-a]pyrazine,  
 [1,2,4]triazolo[1,5-a]pyrazine,  
 [1,2,3]triazolo[4,5-b]pyrazine,  
 [1,2,4]triazolo[4,3-a]quinoxaline,  
 [1,2,3]triazolo[1,5-a]quinoxaline,  
 [1,2,4]triazolo[4,3-b][1,2,4]triazine,  
 [1,2,4]triazolo[3,4-c][1,2,4]triazine,  
 [1,2,4]triazolo[4,3-d][1,2,4]triazine,  
 [1,2,4]triazolo[3,4-f][1,2,4]triazine,  
 [1,2,4]triazolo[1,5-b][1,2,4]triazine,  
 [1,2,4]triazolo[5,1-c][1,2,4]triazine,  
 [1,2,4]triazolo[1,5-d][1,2,4]triazine,  
 [1,2,4]triazolo[4,3-a][1,3,5]triazine,  
 [1,2,4]triazolo[1,5-a][1,3,5]triazine,

tetrazolo[1,5-a]pyridine,  
 tetrazolo[1,5-b]isoquinoline,  
 tetrazolo[1,5-a]quinoline,  
 tetrazolo[5,1-a]isoquinoline,  
 tetrazolo[1,5-b]pyridazine,  
 tetrazolo[1,5-b]quinoline,  
 tetrazolo[5,1-a]phthalazine,  
 tetrazolo[1,5-a]pyrimidine,  
 tetrazolo[1,5-c]pyrimidine,  
 tetrazolo[1,5-a]quinazoline,  
 tetrazolo[1,5-c]quinazoline,  
 tetrazolo[1,5-a]pyrazine,  
 tetrazolo[1,5-a]quinoxaline,  
 tetrazolo[1,5-b][1,2,4]triazine,  
 tetrazolo[5,1-c][1,2,4]triazine,  
 tetrazolo[1,5-d][1,2,4]triazine, and  
 tetrazolo[5,1-f][1,2,4]triazine.

14. (Amended) The multicomponent system of claim 6, wherein component c) [characterized in that as mediators (component c)], as the NO-, NOH- or H-RN-OH-containing compounds [are] , is selected from the group of consisting of cyclical N-hydroxy compounds having at least one optionally substituted 5- or 6-member ring of the structure given in formula A:





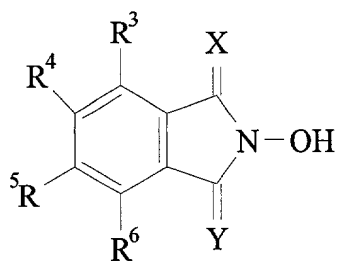
Formula A

as well as their salts, ethers or ester, in which X and Y are the same or different and stand for O, S or NR<sup>1</sup>, in which

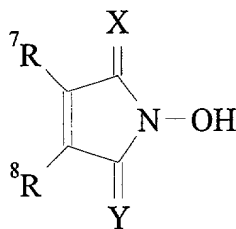
R<sup>1</sup> stands for hydrogen, hydroxyl, formyl, carbamoyl, or sulfono radical, or ester or salt of the sulfono radical, sulfamoyl, nitro, amino, phenyl, aryl C<sub>1</sub>-C<sub>5</sub> alkyl, C<sub>1</sub>-C<sub>12</sub> alkyl, C<sub>1</sub>-C<sub>5</sub> alkoxy, C<sub>1</sub>-C<sub>10</sub> carbonyl, carbonyl C<sub>1</sub>-C<sub>6</sub> alkyl, phospho, phosphono or phosphonooxy radical, or ester or salt of the phosphonooxy radical;

in which carbamoyl, sulfamoyl, amino and phenyl radicals may be unsubstituted or substituted once or multiple times with a radical R<sup>2</sup>, and the aryl C<sub>1</sub>-C<sub>5</sub> alkyl, C<sub>1</sub>-C<sub>12</sub> alkyl, C<sub>1</sub>-C<sub>5</sub> alkoxy, C<sub>1</sub>-C<sub>10</sub> carbonyl, carbonyl C<sub>1</sub>-C<sub>6</sub> alkyl radicals may be saturated or unsaturated, branched or unbranched, and substituted once or multiple times with a radical R<sup>2</sup>, and R<sup>2</sup> is the same or different and stands for hydroxyl, formyl, or carboxyl radical, ester or salt of the carboxyl radical, carbamoyl, sulfono ester or salt of the sulfono radical, sulfamoyl, nitro, amino, phenyl, C<sub>1</sub>-C<sub>5</sub> alkyl, C<sub>1</sub>-C<sub>5</sub> alkoxy radical.

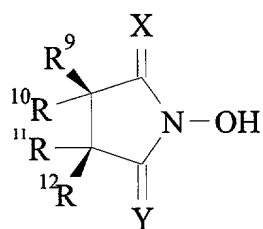
15. (Amended) The multicomponent system of claim 6 [or 14], wherein [characterized in that] as the mediator [(component c)] at least one compound of the general formula VI, VII, VIII or IX is used:



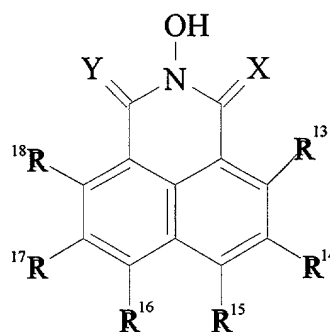
VI



VII



VIII



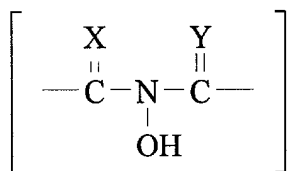
IX

in which X, Y [have the meanings already given] are defined as above and the radicals  $R^3$  to  $R^{18}$  are the same or different and stand for halogen radical, carboxyl radical, salt or ester of a carboxyl radical, or the meaning given for  $R^1$ ;

in which  $R^9$  and  $R^{10}$ , or  $R^{11}$  and  $R^{12}$ , must not at the same time stand for a hydroxyl or amino radical, and

optionally two at a time of the substituents  $R^3$  to  $R^6$ ,  $R^7$  to  $R^8$ ,  $R^9$  to  $R^{12}$ ,  $R^{13}$  to  $R^{18}$  can be linked together into a ring -B-, in which -B- has one of the following meanings:

$(-CH=CH)-_n$ , where  $n = 1-3$ ,  $-CH=CH-CH=N-$ , or



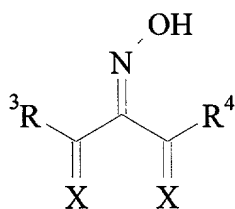
Formula A

and in which optionally the radicals R<sup>9</sup> to R<sup>12</sup> may also be linked to one another by one or two bridge elements -Q-, in which -Q- may be the same or different and can have the following meanings: -O-, -S-, CH<sub>2</sub>-, -CR<sup>19</sup>=CR<sup>20</sup>-;

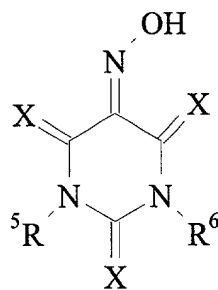
in which R<sup>19</sup> and R<sup>20</sup> are the same or different and have the same meaning as R<sup>3</sup>.

16. (Amended) The multicomponent system of claim 6, [14, 15,] wherein component c) [characterized in that] as the mediator comprises, at least one substance, selected from the group [comprising] consisting of N- hydroxyphthalimide, optionally substituted N- hydroxyphthalimide derivatives, N-hydroxymaleimide, optionally substituted N- hydroxymaleimide derivatives, N- hydroxynaphthalic acid imide, optionally substituted N- hydroxynaphthalic acid imide derivatives, N- hydroxysuccinimide, and optionally substituted N- hydroxysuccinimide derivatives[, is used].

17. (Amended) The multicomponent bleaching system of claim 6, wherein the component c) [characterized in that] as the mediators [(component c),] comprises oxime[s] of the general formula X or XI



X



XI

and their salts, ethers or esters [are used], in which X is the same or different and stands for O, S or NR<sup>1</sup>, in which

R<sup>1</sup> stands for hydrogen, hydroxyl, formyl, carbamoyl, or sulfono radical, or ester or salt of the sulfono radical, sulfamoyl, nitro, amino, phenyl, aryl C<sub>1</sub>-C<sub>5</sub> alkyl, C<sub>1</sub>-C<sub>12</sub> alkyl, C<sub>1</sub>-C<sub>5</sub> alkoxy, C<sub>1</sub>-C<sub>10</sub> carbonyl, carbonyl C<sub>1</sub>-C<sub>6</sub> alkyl, phospho, phosphono or phosphonooxy radical, or ester or salt of the phosphonooxy radical,

in which carbamoyl, sulfamoyl, amino and phenyl radicals may be unsubstituted or substituted once or multiple times with a radical R<sup>2</sup>, and the aryl C<sub>1</sub>-C<sub>5</sub> alkyl, C<sub>1</sub>-C<sub>12</sub> alkyl, C<sub>1</sub>-C<sub>5</sub> alkoxy, C<sub>1</sub>-C<sub>10</sub> carbonyl, carbonyl C<sub>1</sub>-C<sub>6</sub> alkyl radicals may be saturated or unsaturated, branched or unbranched, and substituted once or multiple times with a radical

R<sup>2</sup>, and R<sup>2</sup> is the same or different and stands for hydroxyl, formyl, or carboxyl radical, ester or salt of the carboxyl radical, carbamoyl, sulfono ester or salt of the sulfono radical, sulfamoyl, nitro, amino, phenyl, C<sub>1</sub>-C<sub>5</sub> alkyl, C<sub>1</sub>-C<sub>5</sub> alkoxy radical, and

the radicals R<sup>3</sup> and R<sup>4</sup> are the same or different and stand

for halogen, carboxyl radical, ester or salt of the carboxyl radical, or have the meanings given for R<sup>1</sup>, or are linked together into a ring (-CR<sup>7</sup>R<sup>8</sup>)<sub>n</sub>, where n is equal to 2, 3 or 4, and

R<sup>5</sup> and R<sup>6</sup> have the meanings given for R<sub>1</sub>, and

18. (Amended) The multicomponent bleaching system of claim 6 [one of claims 6 or 17], wherein [characterized in that] as the mediator, compounds of the general formula X, in which X stands for O or S, and the other radicals have the above-given meanings, are used.

20. (Amended) The multicomponent bleaching system of claim 6, wherein [one of claims 6, 17 to 19, characterized in that] as the mediator, alloxane-5-oxime hydrate (violuric acid) or its esters or salts are used as mediators.

17

22. (Amended) The multicomponent system of claim 1 [or 2], wherein component d) comprises [characterized in that as component d), it contains] aliphatic ethers and/or aryl-substituted alcohols[, such as:] selected from the group consisting of 2,3- dimethoxybenzyl alcohol, 3,4- dimethoxybenzyl alcohol, 2,4- dimethoxybenzyl alcohol, 2,6-dimethoxybenzyl alcohol, homovanillyl alcohol, ethylene glycol monophenyl ether, 2- hydroxybenzyl alcohol, 4- hydroxybenzyl alcohol, 4-hydroxy-3- methoxybenzyl alcohol, 2-methoxybenzyl alcohol, 2,5- dimethoxybenzyl alcohol, 2,4-dimethoxybenzylamine, 2,4- dimethoxybenzylamine hydrochloride, veratryl alcohol, and coniferyl alcohol.

23. (Amended) The multicomponent system of claim 1 [or 2], wherein component d) comprises [characterized in that as component (d), it contains] olefins (alkenes)[, such as:] selected from the group consisting of 2-allylphenol, 2-allyl-6-methylphenol, allylbenzene, 3,4- dimethoxypropylbenzene, p-methoxystyrene, 1-allylimidazol, 1-vinylimidazol, styrene, stilbene, allylphenyl ether, cinnamic acid benzyl ester, cinnamic acid methyl ester, 2,4,6- triallyloxy-1,3,5-triazine, 1,2,4- trivinylcyclohexane, 4-allyl-1,2-dimethoxybenzene, 4-tert- butylbenzoic acid vinyl ester, squalene, benzoin allyl ether, cyclohexene, dihydropyran, and N- benzylcinnamic acid anilide.

24. (Amended) The multicomponent system of claim 1 [or 2], wherein component d) comprises [characterized in that as component (d), it contains] phenol ethers[, such as:] selected from the group consisting of 2,3-dimethoxybenzyl alcohol, 3,4- dimethoxybenzyl alcohol, 2,4- dimethoxybenzyl alcohol, 2,6- dimethoxybenzyl alcohol, homovanillyl alcohol, 4- hydroxybenzyl alcohol, 4-hydroxy-3-methoxybenzyl alcohol, 2- methoxybenzyl alcohol, 2,5-

dimethoxybenzyl alcohol, 2,4- dimethoxybenzylamine, 2,4-dimethoxybenzylamine hydrochloride, veratryl alcohol, coniferyl alcohol, veratrol, and anisol.

25. (Amended) The multicomponent system of [claim 1 or 2,] claim 1, wherein component d) comprises [characterized in that as component (d), it contains] carbonyl compounds[, such as:] selected from the group consisting of 4-aminobenzophenone, 4-acetylbiphenyl, benzophenone, benzil, benzophenone hydrazone, 3,4- dimethoxybenzaldehyde, 3,4-dimethoxybenzoic acid, 3,4-dimethoxybenzophenone, 4-dimethylaminobenzaldehyde, 4- acetylbiphenylhydrazone, benzophenone-4-carboxylic acid, benzoyl acetone, bis-(4,4-dimethylamino)benzophenone, benzoin, benzoin oxime, N-benzoyl-N-phenylhydroxylamine, 2- amino-5-chlorobenzophenone, 3-hydroxy-4-methoxybenzaldehyde, 4-methoxybenzaldehyde, anthraquinone-2-sulfonic acid, 4-methylaminobenzaldehyde, benzaldehyde, benzophenone-2- carboxylic acid, 3,3,'4,4'- benzophenonetetracarboxylic acid dianhydride, (S)-(-)-2-(N-benzylpropyl)aminobenzo-henone, benzylphenyl acetic acid anilide, N-benzylbenzanilide, 4,4'- bis-(dimethylamino)-thiobenzophenone, 4,4-bis(diacetyl amino)benzophenone, 2-chlorobenzophenone, 4,4'- dihydroxybenzophenone, 2,4-dihydroxybenzophenone, 3,5- dimethoxy-4-hydroxybenzaldehyde hydrazine, 4- hydroxybenzophenone, 4-methoxybenzophenone, 3,4- dihydroxybenzophenone, p-anisic acid, p-anisic aldehyde, 3,4- dihydroxybenzaldehyde, 3,4-dihydroxybenzoic acid, 3,5-dimethoxy-4-hydroxybenzaldehyde, 3,5-dimethoxy-4- hydroxybenzoic acid, 4-hydroxybenzaldehyde, salicylaldehyde, vanillin, and van[n]ilic acid.

26. (Amended) The multicomponent system of [claims 1 or 2, characterized in that as component (e), it contains] claim 1, wherein the multicomponent system further comprises, as

27. (Amended) The multicomponent system of [claims 1 and 2, characterized in that as oxidoreductases, it contains] claim 3, wherein the oxidoreductases comprise enzymes originating in white rotting fungus [Phanerochaete chrysosporium], Trametes versicolor other fungi, bacteria, animals or plants, which enzymes are obtained from natural organisms or organisms that have been altered by gene technology.

28. (Amended) The multicomponent system of [claims 1 and 2, characterized in that as catalysts it contains] of claim 1, wherein the catalysts comprise modified enzymes, enzyme components, prosthetic groups or mimic substances[, preferably heme groups or compounds containing heme groups].

29. (Amended) The multicomponent system of claim 21, [characterized in that as] wherein the oxidizing agents [it contains] comprise oxygen, which is generated in situ by means of  $H_2O_2$ + catalase or other systems or  $H_2O_2$  from GOD+ glucose or other systems.

30. (Amended) The multicomponent system of claim1, [claims 1 to 29, characterized in that it contains] further comprising cation-forming metal salts.

31. (Amended) The multicomponent system of claim 30, [characterized in that] wherein the cations are  $\text{Fe}^{2+}$ ,  $\text{Fe}^{3+}$ ,  $\text{Mn}^{2+}$ ,  $\text{Mn}^{3+}$ ,  $\text{Mn}^{4+}$ ,  $\text{Cu}^{+}$ ,  $\text{Cu}^{2+}$ ,  $\text{Ti}^{3+}$ ,  $\text{Ce}^{4+}$ ,  $\text{Mg}^{2+}$ , and  $\text{Al}^{3+}$ .



32. (Amended) The multicomponent system of claim 1, [claims 1 and 31, characterized in that in addition it contains] further comprising polysaccharides and/or proteins.

33. (Amended) The multicomponent system of claim 1, [claims 1 to 32, characterized in that as] wherein the polysaccharides [it contains] are selected from the group consisting of glucanes, mannanes, dextrans, levans, pectins, alginates, [or] vegetable rubbers, and[/or] its own polysaccharides formed by the fungi or produced in a mixed culture with yeasts, and as proteins, it contains gelatin or albumin.

34. (Amended) The multicomponent system of claim 1, wherein the additives [claims 1 to 33, characterized in that as the additives it contains] are selected from the group consisting of simple sugar, oligomer sugar, amino acids, polyethylene glycols, polyethylene oxides, polyethylene imines and polydimethyl siloxanes.

35. (Amended) A detergent comprising a multicomponent system wherein the [containing the multicomponent system of one of claims 1 to 34.] multicomponent system comprises

- a) optionally, at least one oxidation catalyst;
- b) at least one suitable oxidizing agent;
- c) at least one mediator, selected from the group [comprising] consisting of hydroxylamines, hydroxylamine derivatives, hydroxamic acids, hydroxamic acid derivatives, and [the] aliphatic, cycloaliphatic, heterocyclic or aromatic compounds that contain at least one N-hydroxy, oxime, N-oxy, or N,N'-dioxy function; and

- d) at least one comediator, selected from the group [comprising] consisting of aryl-substituted alcohols, carbonyl compounds, aliphatic ethers, phenol ethers, and[/or] olefins (alkenes)[; and]  
 [e) optionally, a small quantity of at least one free amine of a respective mediator used].

36. (Amended) [The use of ] A method of using a multicomponent system wherein the multicomponent system comprises [the multicomponent system of one of claims 1 to 35]

- a) optionally, at least one oxidation catalyst;  
 b) at least one suitable oxidizing agent;  
 c) at least one mediator, selected from the group [comprising] consisting of hydroxylamines, hydroxylamine derivatives, hydroxamic acids, hydroxamic acid derivatives, and [the] aliphatic, cycloaliphatic, heterocyclic or aromatic compounds that contain at least one N-hydroxy, oxime, N-oxy, or N,N'-dioxy function; and  
 d) at least one comediator, selected from the group [comprising] consisting of aryl-substituted alcohols, carbonyl compounds, aliphatic ethers, phenol ethers, and[/or] olefins (alkenes); [and]  
 [e) optionally, a small quantity of at least one free amine of a respective mediator used;]  
adding as an additive to detergent formulations with detergent substances or detergent additives known per se.

37. (Amended) The method as claimed in claim 36, wherein the multicomponent system [use of the multicomponent system of one of claims 1 to 36, characterized in that it] is used at a pH value between 2 and 12[, preferably between 4 and 10,] and at a temperature between 10°C and 60°C[, and preferably between 20°C and 40°C].


--38. The multicomponent system as claimed in claim 1, further comprising a predetermined amount of at least one free amine of a mediator.

40. The detergent as claimed in claim 35, wherein the multicomponent system further comprises a predetermined amount of at least one free amine of a mediator.

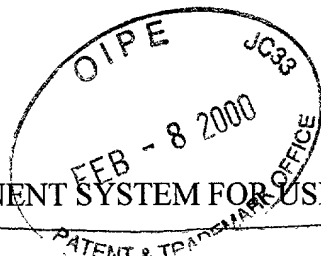
42. The method as claimed in claim 37, wherein the pH value is between 4 and 10.

REMARKS

In view of the foregoing, it is respectfully urged that the present claims are in condition for allowance and reconsideration is requested. An early notice to this effect is earnestly solicited. Should there be any questions regarding this application, the Examiner is invited to contact the undersigned at the number shown below.

  
Karl S. Sawyer, Jr.  
Reg. No. 28,902  
Kennedy, Charleston

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PTO/PCT Rec'd 08 FEB 2000

09/029401

MULTICOMPONENT SYSTEM FOR USE WITH DETERGENT SUBSTANCES

The present invention relates to a novel multicomponent system for use with detergent substances.

5            Particularly in the low-temperature range, conventional bleaching systems in household detergents are unsatisfactory. Below a washing temperature of 60°C, the standard bleaching agent H<sub>2</sub>O<sub>2</sub>/sodium perborate/sodium percarbonate must be activated by adding such chemical bleach activators as TAED (Tetraacetylenediamine) and SNOBS (Sodium p-isononanoyloxy-benzenesulfonate). Moreover, there is a demand for biocompatible bleaching systems with better  
10 biodegradability and that can be used in lower doses for low-temperature clothes washing. While for protein starch and fat dissolution and for fiber treatment in the laundry process enzymes are already in use on an industrial scale, no enzymatic principle has thus far been available for detergent bleaches.

15            In International Patent Disclosure WO 91/05839, the use of various oxidative enzymes (oxidases and peroxidases) have been described for preventing "dye transfer". Peroxidases are known to be capable of bleaching various pigments (3- hydroxyflavon and betaine by horseradish peroxidase, and carotene by peroxidase).

20            The aforementioned patent application describes the decoloration (or bleaching) of textile dyes that have been dissolved out of the laundry and are present in the washwater (conversion of a dyed substrate into an undyed, oxidized substance). The enzyme is intended to have the advantage over hypochlorite, for example, which also attacks the dye or the fabric, of bleaching only dye that is present in dissolved form; hydrogen peroxide or a corresponding precursor, or hydrogen peroxide generated in situ, are involved in the catalysis of the decoloration. The enzyme reaction can be reinforced in part by adding additional oxidizable enzyme substrate, such as metal ions like Mn<sup>++</sup>,

halogen ions such as  $\text{Cl}^-$  or  $\text{Br}^-$ , or organic phenols, such as p-hydroxy cinnamic acid and 2,4-dichlorophenol. Here it is postulated that short-lived radicals or other oxidized states of the added substrate are responsible for the bleaching or other modification of the dyed substance.

US Patent 4,776,768 describes the use of iron porphyrin, haemin chloride, or iron phthalocyanine, or derivatives, together with hydrogen peroxide to prevent dye transfer. However, these substances are rapidly destroyed if there is an excess of peroxide, and the formation of hydrogen peroxide must therefore proceed in controlled fashion.

From International Patent Disclosures WO 94/12619, WO 94/12620 and WO 94/12621, methods are known in which the activity of the peroxidase is enhanced by so called enhancer substances. Such enhancer substances are characterized in terms of their half-life in WO 94/12620. According to WO 94/12621, enhancer substances are characterized by the formula  $\text{A}=\text{N}=\text{N}=\text{B}$ , where A and B are each defined cyclical radicals. According to WO 94/12620, enhancer substances are organic chemicals that contain at least two aromatic rings, at least one of which is substituted with respectively defined radicals.

All three applications pertain to dye transfer inhibition and to the use of the various enhancer substances along with peroxidases as a detergent additive or detergent composition in the laundry detergent field. The combination of these enhancer substances is restricted to peroxidases.

From International Patent Disclosure WO 92/18687, the use of mixtures containing peroxidases is also known. A special system comprising oxidases and substrates suitable for them as well as hydrogen peroxide is described in German Published, Non-Examined Patent Application DE-OS 42 31 761. German Published, Non-Examined Patent Application DE-OS 19 18 729 relates to a further special detergent system, which comprises glucose and glucose oxidase (GOD), or starch, amyloglucosidase and glucose oxidase (GOD) as well as an additive comprising hydroxylamine or

hydroxylamine compounds, in which the hydroxylamine or its derivatives serve to inhibit the catalase that often occurs in GOD.

International Patent Disclosure PCT/EP 94/01967, finally, contains a multicomponent bleaching system for use with detergent substances comprising oxidation catalysts and oxidation agents as well as aliphatic, cycloaliphatic, heterocyclic or aromatic NO-, NOH- or H-NR-OH-containing compounds.

A disadvantage of all the previously known detergent systems is that the cleaning and bleaching action is still unsatisfactory, or that the mediator substances, as in PCT/EP 94/01087, must be added in excessive amounts, possibly leading to environmental and economic problems.

#### General Description of the Invention

The object of the present invention is accordingly to furnish an improved multicomponent system for use with detergents that does not have the above disadvantages of the prior art and that instead increases the action of the actual mediator substances or regenerates in situ, that is, during the washing process.

This object is attained by a multicomponent system, containing

a) optionally, at least one oxidation catalyst;

b) at least one suitable oxidizing agent;

c) at least one mediator, selected from the group comprising hydroxylamines, hydroxylamine derivatives, hydroxamic acids, hydroxamic acid derivatives, and the aliphatic, cycloaliphatic, heterocyclic or aromatic compounds that contain at least one N-hydroxy, oxime, N-oxy, or N,N'-dioxy function;

d) at least one comediator, selected from the group comprising aryl-substituted alcohols, carbonyl compounds, aliphatic ethers, phenol ethers, and/or olefins (alkenes); and

e) optionally, a small quantity of at least one free amine of a respective mediator used.

It is surprising that when the aforementioned comediators are added to the aforementioned mediators, optionally together with the free amines of the respective mediators and oxidation catalysts, on the one hand the bleaching action of detergents can be improved considerably, and on the other the consumption of mediators can be reduced.

According to the invention, either one or more of the aforementioned mediators and comediators can be used. It is preferable to use one mediator and one comediator. It is also conceivable to use one mediator and two or more comediators. Conversely, it is also possible to use two or more mediators with one comediator.

The substances in the multicomponent bleaching system listed in paragraphs a), b), c), d), and e) are preferably used in a ratio of 2:0.2:10:0.2:0.2, and each component in the system can be multiplied by from 2 to 10.

The individual components of the multicomponent system according to the invention will be described in further detail below:

#### Oxidation Catalysts

The multicomponent system of the invention preferably contains at least one oxidation catalyst. Enzymes are preferably used as the oxidation catalysts. The term "enzyme" also includes enzymatically active proteins or peptides or prosthetic groups of enzymes.

As the enzyme, in the multicomponent system of the invention, oxidoreductases of classes 1.1.1 through 1.97 according to the International Enzyme Nomenclature, Committee of the International Union of Biochemistry and Molecular Biology (Enzyme Nomenclature, Academic Press, Inc., 1992, pp. 24-154), can be used.

Preferably, enzymes of the following named classes are used:



Enzymes of class 1.1, which include all the dehydrogenases that act on primary and secondary alcohols and semiacetals, and which as acceptors have  $\text{NAD}^+$  or  $\text{NADP}^+$  (subclass 1.1.1.), cytochromes (1.1.2), oxygen ( $\text{O}_2$ ) (1.1.3), disulfides (1.1.4), quinones (1.1.5), or other acceptors (1.1.99). From this class, the enzymes of class 1.1.5 with quinones as acceptors and the enzymes of class 1.1.3. with oxygen as an acceptor are especially preferred and in this latter class cellobiose: quinone-1-oxidoreductase (1.1.5.1) is particularly preferred.

Enzymes of class 1.2 can also be used. This enzyme class (1.1.5.1) includes enzymes of the type that oxidize aldehydes to form the corresponding acids or oxo groups. The acceptors may be  $\text{NAD}^+$ ,  $\text{NADP}^+$  (1.2.1.), cytochromes (1.2.2), oxygen (1.2.3), sulfides (1.2.4), iron-sulfur proteins (1.2.5), or other acceptors (1.2.99). Here, the enzymes of group (1.2.3) with oxygen as the acceptor are especially preferred.

Enzymes of class 1.3 can also be used. This class summarizes enzymes that act on  $\text{CH-CH}$  groups of the donor. The corresponding acceptors are  $\text{NAD}^+$ ,  $\text{NADP}^+$  (1.3.1), cytochromes (1.3.2), oxygen (1.3.3), quinones or related compounds (1.3.5), iron-sulfur proteins (1.3.7), or other acceptors (1.3.99). Once again, the enzymes of class (1.3.3) with oxygen as an acceptor and (1.3.5) with quinones etc. as an acceptor are especially preferred.

The enzymes of class 1.4 that act on  $\text{CH-NH}_2$  groups of the donor can also be used. The corresponding acceptors are  $\text{NAD}^+$ ,  $\text{NADP}^+$  (1.4.1.), cytochromes (1.4.2), oxygen (1.4.3), disulfides (1.4.4), iron-sulfur proteins (1.4.7), or other acceptors (1.4.99) Once again, enzymes of class 1.4.3 with oxygen as an acceptor are especially preferred.

Also usable are enzymes of class 1.5 that act on  $\text{CH-NH}$  groups of the donor. The corresponding acceptors are  $\text{NAD}^+$ ,  $\text{NADP}^+$  (1.5.1), oxygen (1.4.3), disulfides (1.5.4), quinones

(1.5.5), or other acceptors (1.5.99). Once again, enzymes with oxygen ( $O_2$ ) (1.5.3) and with quinones (1.5.5) as acceptors are especially preferred.

Enzymes of class 1.6 that act on NADH or NADPH can also be used. In this case the acceptors are  $NADP^+$  (1.6.1), heme proteins (1.6.2), disulfides (1.6.4), quinones (1.6.5),  $NO_2$  groups (1.6.6), and a flavin (1.6.8) or some other acceptors (1.6.99). Enzymes of class 1.6.5 with quinones as the acceptors are especially preferred here.

Furthermore, enzymes of class 1.7 that act on other  $NO_2$  compounds as donors and that as acceptors have cytochromes (1.7.2), oxygen ( $O_2$ ) (1.7.3), iron-sulfur proteins (1.7.7) or others (1.7.99) can also be used. Here, class 1.7.3 with oxygen as the acceptor is especially preferred.

Enzymes of class 1.8 that act on sulfur groups as donors and that as acceptors have  $NAD^+$ ,  $NADP^+$  (1.8.1) cytochromes (1.8.2), oxygen ( $O_2$ ) (1.8.3), disulfides (1.8.4), quinones (1.8.5), iron-sulfur proteins (1.8.7), or others (1.8.99) can also be used. Class 1.8.3 with oxygen ( $O_2$ ) and (1.8.5) with quinones as acceptors is especially preferred.

Also usable are enzymes of class 1.9, which act on heme groups as donors and which have as acceptors oxygen ( $O_2$ ) (1.9.3),  $NO_2$  compounds (1.9.6) and others (1.9.99). Here, group 1.9.3 with oxygen ( $O_2$ ) as an acceptor (cytochromooxidases) is especially preferred here.

Moreover, enzymes of class 1.12 that act on hydrogen as a donor can be considered. The acceptors are  $NAD^+$  or  $NADP^+$  (1.12.1) or others (1.12.99).

The enzymes that can be used also include those of classes 1.13 and 1.14 (oxygenases).

Enzymes of class 1.15, which act on superoxide radicals as acceptors, can also be named. Here, the superoxide dismutase (1.15.1.1) is especially preferred.

In addition, enzymes of class 1.16 can be used.  $NAD^+$  or  $NADP^+$  (1.16.1) or oxygen ( $O_2$ ) (1.16.3) act as acceptors. Especially preferred here are enzymes of class 1.16.3.1

(ferroxidases, such as ceruloplasmin). Others that can be named are those enzymes that belong to group 1.17 (action on  $\text{CH}_2$  groups which are oxidized to form  $-\text{CHOH}-$ ), 1.18 (action on reduced ferredoxin as a donor), 1.19 (action on reduced flavodoxin as a donor), and 1.97 (other oxidoreductases).

5           Among the enzymes that are very particularly preferred are those in class 1.10 that act on biphenols and related compounds. They catalyze the oxidation of biphenols and ascorbates. Functioning as acceptors are  $\text{NAD}^+$ ,  $\text{NADP}^+$  (1.10.1), cytochromes (1.10.2), oxygen (1.10.3), or others (1.10.99). Of these, again, enzymes of class 1.10.3 with oxygen ( $\text{O}_2$ ) as the acceptor are especially preferred.

10           Of the enzymes of this class, the enzymes catechol oxidase (tyrosinase) (1.10.3.1), L-ascorbate oxidase (1.10.3.3), O-aminophenol oxidase (1.10.3.4) and laccase (benzenediol: oxygen oxidoreductase) (1.10.3.2); the laccases (benzenediol: oxygen oxidoreductase) (1.10.3.2) are especially preferred.

15           Also particularly preferred are the enzymes of group 1.11 that act on a peroxide as the acceptor. This single subclass (1.11.1) contains the peroxidases.

          Also especially preferred are the enzymes of group 1.11., which act on a peroxide as an acceptor.

20           This single subclass (1.11.1) includes the peroxidases. Very particularly preferred here are the cytochrome C peroxidases (1.11.1.5), catalase (1.11.1.6), peroxidase (1.11.1.7), iodide peroxidase (1.11.1.8), glutathione peroxidase (1.11.1.9), chloride peroxidase (1.11.1.10), L-ascorbate peroxidase (1.11.1.11), phospholipid hydroperoxide glutathione peroxidase (1.11.1.12), manganese peroxidase (1.12.1.13), diarylpropane peroxidase (ligninase, lignin peroxidase).

These enzymes are commercially available or can be prepared by standard methods.

Organisms that can be considered for producing the enzymes are for instance plants, animal cells, bacteria, and fungi. In principle, both naturally occurring organisms and organisms altered by gene technology can be enzyme producers. Parts of singlecell or multicell organisms, and above all cell cultures, are also conceivable as enzyme producers.

Particular for producing the preferred enzymes of group 1.11.1, but above all from group 1.10.3, and especially for producing the laccases, white rotting fungi such as Pleurotus, Phlebia and Trametes, for instance, are used.

#### Oxidation Agent

The multicomponent system of the invention includes at least one oxidation agent. As the oxidizing agent, air, oxygen, ozone,  $H_2O_2$ , organic peroxides, peracids such as peracetic acid, performic acid, persulfuric acid, pernitric acid, metachloroperoxybenzoic acid, perchloric acid, perborates, peracetates, persulfates, peroxides, or oxygen species and their free radicals thereof, such as OH, OOH and superoxide ( $O_2^-$ ) and singlet oxygen, ozonide, dioxygenyl cation ( $O_2^+$ ), dioxiranes, dioxitanes, or Fremy radicals can for instance be used.

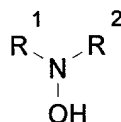
The oxidation agents used are preferably those that can either be generated by the corresponding oxidoreductases, such as dioxiranes from laccases plus carbonyls, or that can chemically regenerate the mediator (for instance, Caro's acid + benzotriazole produces hydroxybenzotriazole) or that can convert these directly.

#### Mediators

The multicomponent system according to the invention contains as a mediator (component c) preferably at least one compound that contains an N-hydroxy, oxime, N-oxy or D-dioxy function and/or one of the compounds named below of formulas I, II, III, IV or V; the compounds of

formulas II, III, IV and V are preferred, the compounds of formulas II, IV and V are especially preferred, and compounds of formulas IV and V are very particularly preferred.

Hydroxylamines can for instance be used according to the invention (open-chained or cyclical, aliphatic or aromatic, heterocyclical), of the general formula



in which the substituents R<sup>1</sup> and R<sup>2</sup>, which may be the same or different, independently of one another represent one of the following groups: hydrogen, C<sub>1</sub>-C<sub>12</sub> alkyl, carbonyl C<sub>1</sub>-C<sub>6</sub> alkyl, phenyl, aryl, of which C<sub>1</sub>-C<sub>12</sub> alkyl, carbonyl C<sub>1</sub>-C<sub>6</sub> alkyl, phenyl, aryl groups may be unsubstituted or may also be substituted once or multiple times with the radical R<sup>3</sup>.

The radical R<sup>3</sup> may represent one of the following groups: hydrogen, halogen, hydroxyl, formyl, carboxy and salts and esters thereof; amino, nitro, C<sub>1</sub>-C<sub>12</sub> alkyl, C<sub>1</sub>-C<sub>6</sub> alkyloxy, carbonyl C<sub>1</sub>-C<sub>6</sub> alkyl, phenyl, sulfono, their esters and salts, sulfamoyl, carbamoyl, phospho, phosphono, phosphonooxy and their salts and esters. The amino, carbamoyl and sulfamoyl groups of the radical R<sup>3</sup> may be unsubstituted or may be substituted once or two times with hydroxy, C<sub>1</sub>-C<sub>3</sub> alkyl, C<sub>1</sub>-C<sub>3</sub> alkoxy.

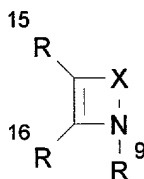
The radicals R<sup>1</sup> and R<sup>2</sup> can jointly form a group -B-. In that case, -B- represents one of the following groups: (-CHR<sup>4</sup>)<sub>n</sub>, (CR<sup>4</sup>=CH-)<sub>m</sub>. n represents an integer from 1 to 6 and m represents an integer from 1 to 3.

R<sup>4</sup> is a substituent that is defined like R<sup>3</sup>.

Examples of hydroxylamines that can be used are: N,N- dipropylhydroxylamine, N,N-diisopropylhydroxylamine, N- hydroxypyrrolidine, N-hydroxypiperidine, N-

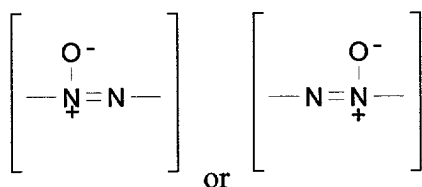
hydroxyhexahydroazepine, N,N-dibenzylhydroxylamine, phenylhydroxylamine, 3-hydroxylamino-3-phenyl propionic acid, 2-hydroxylamino-3-phenyl propionic acid, N- sulfomethylhydroxylamine, N-sulfomethylhydroxylamine.

Compounds of general formula II are:



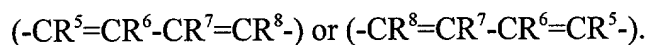
5

X stands for one of the following groups:  $(-N=N-)$ ,  $(-N=CR^{10}-)_p$ ,  $(-CR^{10}=N-)_p$ ,  $(-CR^{11}=CR^{12}-)_p$ ,



and p is equal to 1 or 2.

The radicals  $R^9$  to  $R^{12}$ ,  $R^{15}$  and  $R^{16}$  may be the same or different and independently of one another can represent one of the following groups: hydrogen, halogen, hydroxyl, formyl, carboxyl and salts and esters thereof, amino, nitro,  $C_1$ - $C_{12}$  alkyl,  $C_1$ - $C_6$  alkyloxy, carbonyl  $C_1$ - $C_6$  alkyl, phenyl, sulfono, esters and salts thereof, sulfamoyl, carbamoyl, phospho, phosphono, phosphonoxy and their salts and esters. The amino, carbamoyl and sulfamoyl groups of the radicals  $R^9$  to  $R^{12}$ ,  $R^{15}$  and  $R^{16}$  may be unsubstituted or may also be substituted once or two times with hydroxyl,  $C_1$ - $C_3$  alkyl,  $C_1$ - $C_3$  alkoxy. The radicals  $R^{15}$  and  $R^{16}$  can form a common group -G-. -G- represents one of the following groups:



The radicals  $R^5$  to  $R^8$  may be the same or different and independently of one another can represent one of the following groups: hydrogen, halogen, hydroxyl, formyl, carboxyl and salts and esters thereof; amino, nitro,  $C_1$ - $C_{12}$  alkyl,  $C_1$ - $C_6$  alkyloxy, carbonyl,  $C_1$ - $C_6$  alkyl, phenyl, sulfono, esters and salts thereof, sulfamoyl, carbamoyl, phospho, phosphono, phosphonooxy and their salts and esters. The amino, carbamoyl and sulfamoyl groups of the radicals  $R^5$  to  $R^8$  may be unsubstituted or may also be substituted once or two times with hydroxy,  $C_1$ - $C_3$  alkyl,  $C_1$ - $C_3$  alkoxy.

The  $C_1$ - $C_{12}$  alkyl,  $C_1$ - $C_6$  alkyloxy, carbonyl  $C_1$ - $C_6$  alkyl, phenyl and aryl groups of radicals  $R^5$  to  $R^8$  may be unsubstituted or may be substituted one or two times with the radical  $R^{18}$ .

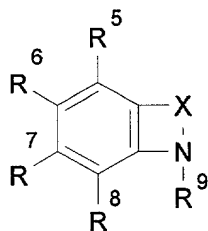
The radical  $R^{18}$  can represent one of the following groups: hydrogen, halogen, hydroxyl, formyl, carboxyl and their salts and esters, amino, nitro,  $C_1$ - $C_{12}$  alkyl,  $C_1$ - $C_6$  alkyloxy, carbonyl  $C_1$ - $C_6$  alkyl, phenyl, aryl, and their esters and salts. The carbamoyl, sulfamoyl and amino groups of the radical  $R^{18}$  may be unsubstituted or may also be substituted once or two times with the radical  $R^{19}$ .

The radical  $R^{19}$  may represent one of the following groups: hydrogen; hydroxyl, formyl, carboxyl and their salts and esters; amino, nitro,  $C_1$ - $C_{12}$  alkyl,  $C_1$ - $C_6$  alkyloxy, carbonyl  $C_1$ - $C_6$  alkyl, phenyl and aryl.

Examples of the above-named compounds are

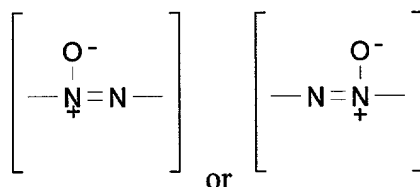
1-hydroxy-1,2,3-triazole-4,5-dicarboxylic acid, 1-phenyl-1H-1,2,3-triazole-3-oxide, 5-chloro-1-phenyl-1H-1,2,3-triazole-3-oxide, 5-methyl-1-phenyl-1H-1,2,3-triazole-3-oxide, 4-(2,2-dimethylpropanoyl)-1-hydroxy-1H-1,2,3-triazole, 4-hydroxy-2-phenyl-2H-1,2,3-triazole-1-oxide, 2,4,5-triphenyl-2H-1,2,3-triazole-1-oxide, 1-benzyl-1H-1,2,3-triazole-3-oxide, 1-benzyl-4-chloro-1H-1,2,3-triazole-3-oxide, 1-benzyl-4-bromo-1H-1,2,3-triazole-3-oxide, and 1-benzyl-4-methoxy-1H-1,2,3-triazole-3-oxide.

Compounds of the general structure III are:



X stands for one of the following groups:  $(-\text{N}=\text{N}-)$ ,  $(-\text{N}=\text{CR}^{10}-)_p$ ,  $(-\text{CR}^{10}=\text{N}-)_p$ ,

5  $(-\text{CR}^{11}=\text{CR}^{12}-)_p$



and p is equal to 1 or 2.

The radicals  $\text{R}^5$  to  $\text{R}^{12}$  may be the same or different and independently of one another can represent one of the following groups: hydrogen, halogen, hydroxyl, formyl, carboxyl and salts and esters thereof, amino, nitro,  $\text{C}_1$ - $\text{C}_{12}$  alkyl,  $\text{C}_1$ - $\text{C}_6$  alkyloxy, carbonyl  $\text{C}_1$ - $\text{C}_6$  alkyl, phenyl, aryl, sulfono esters and salts thereof, sulfamoyl, carbamoyl, phospho, phosphono, phosphonooxy and their salts and esters. The amino, carbamoyl and sulfamoyl groups of the radicals  $\text{R}^5$  to  $\text{R}^{12}$  may be unsubstituted or may also be substituted once or two times with hydroxyl,  $\text{C}_1$ - $\text{C}_3$  alkyl,  $\text{C}_1$ - $\text{C}_3$  alkoxy. The  $\text{C}_1$ - $\text{C}_{12}$  alkyl,  $\text{C}_1$ - $\text{C}_6$  alkyloxy, carbonyl  $\text{C}_1$ - $\text{C}_6$  alkyl, phenyl, aryl, aryl  $\text{C}_1$ - $\text{C}_6$  alkyl groups of radicals  $\text{R}^5$  to  $\text{R}^{12}$  may be unsubstituted or substituted one or two times with the radical  $\text{R}^{13}$ .

The radical  $\text{R}^{13}$  can represent one of the following groups: hydrogen, halogen, hydroxyl, formyl, carboxyl and their salts and esters; amino, nitro,  $\text{C}_1$ - $\text{C}_{12}$  alkyl,  $\text{C}_1$ - $\text{C}_6$  alkyloxy, carbonyl  $\text{C}_1$ - $\text{C}_6$  alkyl, phenyl, aryl, sulfono, sulfeno, sulfino, and their esters and salts. The carbamoyl, sulfamoyl,

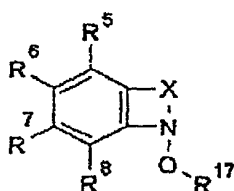


amino groups of the radical  $R^{13}$  may be unsubstituted or may also be substituted once or two times with the radical  $R^{14}$ .

The radical  $R^{14}$  may represent one of the following groups: hydrogen; hydroxyl, formyl, carboxyl and their salts and esters; amino, nitro,  $C_1$ - $C_{12}$  alkyl,  $C_1$ - $C_6$  alkyloxy, carbonyl  $C_1$ - $C_6$  alkyl, phenyl or aryl.

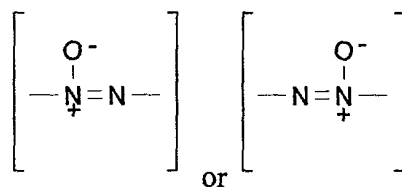
Examples are: 1-hydroxy-benzimidazoles, such as 1-hydroxy-benzimidazole-2-carboxylic acid, 1-hydroxy-benzimidazole, 2-methyl-1-hydroxy-benzimidazole, 2-phenyl-1-hydroxy-benzimidazole, and 1-hydroxyindoles, such as 2-phenyl-1-hydroxyindole.

Substances of general formula IV are:



X stands for one of the following groups:  $(-N=N-)$ ,  $(-N=CR^{10}-)_m$ ,  $(-CR^{10}=N-)_m$ ,

$(-CR^{11}=CR^{12}-)_m$

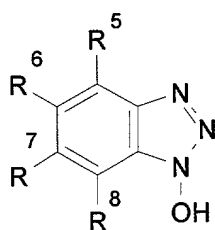


and m is equal to 1 or 2.

For the radicals  $R^5$  to  $R^8$  and  $R^{10}$  to  $R^{12}$  what has been said above applies.

$R^{17}$  can be hydrogen,  $C_1$ - $C_{10}$  alkyl,  $C_1$ - $C_{10}$  carbonyl, of which  $C_1$ - $C_{10}$  alkyl and  $C_1$ - $C_{10}$  carbonyl can be unsubstituted or mono- or polysubstituted with a radical  $R^{18}$ , which is defined like  $R^3$ .

5 Of the substances of Formula IV, in particular derivatives of 1-hydroxybenzotriazole and of the tautomeric benzotriazole-1-oxide, as well as their esters and salts, are preferred (compounds of formula V):



10 The radicals  $R^5$  to  $R^8$  may be the same or different and independently of one another can represent one of the following groups: hydrogen, halogen, hydroxyl, formyl, carboxyl and salts and esters thereof; amino, nitro,  $C_1$ - $C_{12}$  alkyl,  $C_1$ - $C_6$  alkyloxy, carbonyl  $C_1$ - $C_6$  alkyl, phenyl, sulfono esters and salts thereof, sulfamoyl, carbamoyl, phospho, phosphono, phosphonooxy and their salts and esters. The amino, carbamoyl and sulfamoyl groups of the radicals  $R^5$  to  $R^8$  may be  
 15 unsubstituted or may also be substituted once or two times with hydroxyl,  $C_1$ - $C_3$  alkyl,  $C_1$ - $C_3$  alkoxy. The  $C_1$ - $C_{12}$  alkyl,  $C_1$ - $C_6$  alkyloxy, carbonyl  $C_1$ - $C_6$  alkyl, phenyl or aryl groups of radicals  $R^5$  to  $R^8$  may be unsubstituted or may also be substituted one or mono- or polysubstituted with the radical  $R^{18}$ .

20 The radical  $R^{18}$  can represent one of the following groups: hydrogen, halogen, hydroxyl, formyl, carboxyl and their salts and esters; amino, nitro,  $C_1$ - $C_{12}$  alkyl,  $C_1$ - $C_6$  alkyloxy, carbonyl  $C_1$ - $C_6$  alkyl, phenyl, aryl, sulfono, sulfeno, sulfino, and their esters and salts. The carbamoyl, sulfamoyl, amino groups of the radical  $R^{18}$  may be unsubstituted or may also be substituted once or two times

with the radical R<sup>19</sup>. The radical R<sup>19</sup> may represent one of the following groups: hydrogen; hydroxyl, formyl, carboxyl and their salts and esters, amino, nitro, C<sub>1</sub>-C<sub>12</sub> alkyl, C<sub>1</sub>- C<sub>6</sub> alkyloxy, carbonyl C<sub>1</sub>-C<sub>6</sub> alkyl, phenyl or aryl.

Examples of the compounds named are 1-H- hydroxybenzotriazoles, such as: 1-

- 5 hydroxybenzotriazole, 1- hydroxybenzotriazole-6-sulfonic acid, 1-hydroxybenzotriazole- 6-carboxylic acid, 1-hydroxybenzotriazole-6-N- phenylcarboxamide, 5-ethoxy-6-nitro-1-hydroxybenzotriazole, 4-ethyl-7-methyl-6-nitro-1-hydroxybenzotriazole, 2,3-bis-(4- ethoxyphenyl)-4,6-dinitro-2,3-dihydro-1-hydroxybenzotriazole, 2,3-bis-(2-bromo-4-methylphenyl)-4,6-dinitro-2,3-dihydro-1- hydroxybenzotriazole, 2,3-bis-(4-bromophenyl)-4,6-dinitro- 2,3-dihydro-1-hydroxybenzotriazole, 2,3-bis-(4- carboxyphenyl)-4,6-dinitro-2,3-dihydro-1- hydroxybenzotriazole, 4,6-bis-(trifluoromethyl)-1- hydroxybenzotriazole, 5-bromo-1-hydroxybenzotriazole, 6- bromo-1-hydroxybenzotriazole, 4-bromo-7-methyl-1- hydroxybenzotriazole, 5-bromo-7-methyl-6-nitro-1-hydroxybenzotriazole, 4-bromo-6-nitro-1-hydroxybenzotriazole, 6-bromo-4-nitro-1-hydroxybenzotriazole, 4-chloro-1- hydroxybenzotriazole, 6-chloro-5-isopropyl-1-hydroxybenzotriazole, 5-chloro-6-methyl-1- hydroxybenzotriazole, 6-chloro-5-methyl-1-hydroxybenzotriazole, 4-chloro-7-methyl-6-nitro-1- hydroxybenzotriazole, 5-chloro-1-hydroxybenzotriazole, 6- chloro-1-hydroxybenzotriazole, 4-chloro-5-methyl-1-hydroxybenzotriazole, 5-chloro-4-methyl-1- hydroxybenzotriazole, 4-chloro-6-nitro-1-hydroxybenzotriazole, 6-chloro-4-nitro-1- hydroxybenzotriazole, 7-chloro-1-hydroxybenzotriazole, 6- diacetylamino-1-hydroxybenzotriazole, 2,3-dibenzyl-4,6- dinitro-2,3-dihydro-1-hydroxybenzotriazole, 4,6-dibromo-1- hydroxybenzotriazole, 4,6-dichloro-1-hydroxybenzotriazole, 5,6-dichloro-1-hydroxybenzotriazole, 4,5-dichloro-1-hydroxybenzotriazole, 4,7-dichloro-1-hydroxybenzotriazole, 5,7-dichloro-6-nitro-1-hydroxybenzotriazole, 5,6-dimethoxy-1-

hydroxybenzotriazole, 2,3-di[2]naphthyl-4,6-dinitro-2,3- dihydro-1-hydroxybenzotriazole, 4,6-  
dinitro-1- hydroxybenzotriazole, 4,6-dinitro-2,3-diphenyl-2,3-dihydro-1- hydroxybenzotriazole, 4,6-  
dinitro-2,3-di-p-tolyl-2,3-dihydro- 1-hydroxybenzotriazole, 5-hydrazino-7-methyl-4-nitro-1-  
hydroxybenzotriazole, 5,6-dimethyl-1-hydroxybenzotriazole, 4- methyl-1-hydroxybenzotriazole, 5-  
5 methyl-1- hydroxybenzotriazole, 6-methyl-1-hydroxybenzotriazole, 5-(1- methylethyl)-1-  
hydroxybenzotriazole, 4-methyl-6-nitro-1- hydroxybenzotriazole, 6-methyl-4-nitro-1-  
hydroxybenzotriazole, 5-methoxy-1-hydroxybenzotriazole, 6- methoxy-1-hydroxybenzotriazole, 7-  
methyl-6-nitro-1- hydroxybenzotriazole, 4-nitro-1-hydroxybenzotriazole, 6- nitro-1-  
hydroxybenzotriazole, 6-nitro-4-phenyl-1- hydroxybenzotriazole, 5-phenylmethyl-1-  
10 hydroxybenzotriazole, 4-trifluoromethyl-1-hydroxybenzotriazole, 5-trifluoromethyl- 1-  
hydroxybenzotriazole, 6-trifluoromethyl-1- hydroxybenzotriazole, 4,5,6,7-tetrachloro-1-  
hydroxybenzotriazole, 4,5,6,7-tetrafluoro-1- hydroxybenzotriazole, 6-tetrafluoroethyl-1-  
hydroxybenzotriazole, 4,5,6-trichloro-1-hydroxybenzotriazole, 4,6,7-trichloro-1-  
hydroxybenzotriazole, 6-sulfamido-1- hydroxybenzotriazole, 6-N,N-diethylsulfamido-1-  
15 hydroxybenzotriazole, 6-N-methylsulfamido-1- hydroxybenzotriazole, 6-(1H-1,2,4-triazole-1-  
ylmethyl)-1- hydroxybenzotriazole, 6-(5,6,7,8-tetrahydroimidazo-[1,5- a]pyridin-5-yl)-1-  
hydroxybenzotriazole, 6-(phenyl-1H-1,2,4- triazole-1-ylmethyl)-1-hydroxybenzotriazole, 6-[(5-  
methyl-1H- imidazole-1-yl)phenylmethyl]-1-hydroxybenzotriazole, 6-[(4-  
methyl-1H-imidazole-1-yl)phenylmethyl]-1- hydroxybenzotriazole, 6-[(2-methyl-1H-imidazole-1-  
20 yl)phenylmethyl]-1-hydroxybenzotriazole, 6-(1H-imidazole-1- ylphenylmethyl)-1-  
hydroxybenzotriazole, 5-(1H-imidazole-1- ylphenylmethyl)-1-hydroxybenzotriazole, 6-[1(1H-  
imidazole-1- yl)ethyl]-1-hydroxybenzotriazole monohydrochloride.

In this connection, the following salts should also be mentioned:

1-hydroxybenzotriazole, sodium salt.

1-hydroxybenzotriazole, potassium salt.

1-hydroxybenzotriazole, lithium salt.

1-hydroxybenzotriazole, ammonium salt.

5 1-hydroxybenzotriazole, calcium salt.

1-hydroxybenzotriazole, magnesium salt.

1-hydroxybenzotriazole-6-sulfonic acid, monosodium salt.

Other examples of compounds of formulas IV and V that can be used according to the invention are 3H-benzotriazole-1-oxides, such as: 3H-benzotriazole-1-oxide, 6-acetyl-3H-benzotriazole-1-oxide, 5-ethoxy-6-nitro-3H-benzotriazole-1-oxide, 4-ethyl-7-methyl-6-nitro-3H-benzotriazole-1-oxide, 6-amino-3,5-dimethyl-3H-benzotriazole-1-oxide, 6-amino-3-methyl-3H-benzotriazole-1-oxide, 5-bromo-3H-benzotriazole-1-oxide, 6-bromo-3H-benzotriazole-1-oxide, 4-bromo-7-methyl-3H-benzotriazole-1-oxide, 5-bromo-4-chloro-6-nitro-3H-benzotriazole-1-oxide, 4-bromo-6-nitro-3H-benzotriazole-1-oxide, 6-bromo-4-nitro-3H-benzotriazole-1-oxide, 5-chloro-3H-benzotriazole-1-oxide, 6-chloro-3H-benzotriazole-1-oxide, 4-chloro-6-nitro-3H-benzotriazole-1-oxide, 4,6-dibromo-3H-benzotriazole-1-oxide, 4,6-dibromo-3-methyl-3H-benzotriazole-1-oxide, 4,6-dichloro-3H-benzotriazole-1-oxide, 4,7-dichloro-3H-benzotriazole-1-oxide, 5,6-dichloro-3H-benzotriazole-1-oxide, 4,6-dichloro-3-methyl-3H-benzotriazole-1-oxide, 5,7-dichloro-6-nitro-3H-benzotriazole-1-oxide, 3,6-dimethyl-6-nitro-3H-benzotriazole-1-oxide, 3,5-dimethyl-6-nitro-3H-benzotriazole-1-oxide, 3-methyl-3H-benzotriazole-1-oxide, 5-methyl-3H-benzotriazole-1-oxide, 6-methyl-3H-benzotriazole-1-oxide, 6-methyl-4-nitro-3H-benzotriazole-1-oxide, 7-methyl-6-nitro-3H-benzotriazole-1-oxide, 5-chloro-6-nitro-3H-benzotriazole-1-oxide.

Further examples of compounds of formulas IV and V are 2H-benzotriazole-1-oxides, such as:

(2-(4-acetoxyphenyl)-2H-benzotriazole-1-oxide, 6- acetylamino-2-phenyl-2H-benzotriazole-1-oxide, 2-(4- ethylphenyl)-4,6-dinitro-2H-benzotriazole-1-oxide, 2-(3- aminophenyl)-2H-benzotriazole-1-oxide, 2-(4-aminophenyl)-2H- benzotriazole-1-oxide, 6-amino-2-phenyl-2H-benzotriazole-1- oxide, 5-bromo-4-chloro-6-nitro-2H-benzotriazole-1-oxide, 2- (4-bromophenyl)-2H-benzotriazole-1-oxide, 5-bromo-2-phenyl- 2H-benzotriazole-1-oxide, 6-bromo-2-phenyl-2H-benzotriazole- 1-oxide, 2-(4-bromophenyl)-4,6-dinitro-2H-benzotriazole-1- oxide, 2-(4-bromophenyl)-6-nitro-2H-benzotriazole-1-oxide, 5- chloro-2-(2-chlorophenyl)-2H-benzotriazole-1-oxide, 5-chloro- 2-(3-chlorophenyl)-2H-benzotriazole-1-oxide, 5-chloro-2-(2,4- dibromophenyl)-2H-benzotriazole-1-oxide, 5-chloro-2-(2,5- dimethylphenyl)-2H-benzotriazole-1-oxide, 5-chloro-2-(4- nitrophenyl)-2H-benzotriazole-1-oxide, 5-chloro-6-nitro-2H-benzotriazole-1-oxide, 2-[4-(4-chloro-3-nitrophenylazo)-3- nitrophenyl]-4,6-dinitro-2H-benzotriazole-1-oxide, 2-(3-chloro-4-nitrophenyl)-4,6-dinitro-2H-benzotriazole-1-oxide, 2-(4-chloro-3-nitrophenyl)-4,6-dinitro-2H-benzotriazole-1-oxide, 4-chloro-6-nitro-2-p-tolyl-2H-benzotriazole-1-oxide, 5-chloro-6-nitro-2-p-tolyl-2H-benzotriazole-1-oxide, 6- chloro-4-nitro-2-p-tolyl-2H-benzotriazole-1-oxide, 2-(2- chlorophenyl)-2H-benzotriazole-1-oxide, 2-(3-chlorophenyl)- 2H-benzotriazole-1-oxide, 2-(4-chlorophenyl)-2H-benzotriazole-1-oxide, 5-chloro-2-phenyl-2H-benzotriazole-1- oxide, 2-[4-(4-chlorophenylazo)-3-nitrophenyl]-4,6-dinitro- 2H-benzotriazole-1-oxide, 2-(2-chlorophenyl)-4,6-dinitro-2H-benzotriazole-1-oxide, 2-(3-chlorophenyl)-4,6-dinitro-2H- benzotriazole-1-oxide, 2-(4-chlorophenyl)-4,6-dinitro-2H- benzotriazole-1-oxide, 2-{4-[N'-(3-chlorophenyl)hydrazino]-3-nitrophenyl}-4,6-dinitro-2H-benzotriazole-1-oxide, 2-{4-[N'-(4-Chlorophenyl)hydrazino]-3-nitrophenyl}-4,6-dinitro-2H- benzotriazole-1-oxide, 2-(2-chlorophenyl)-6-methyl-2H- benzotriazole-

1-oxide, 2-(3-chlorophenyl)-6-methyl-2H- benzotriazole-1-oxide, 2-(4-chlorophenyl)-6-methyl-2H-  
 benzotriazole-1-oxide, 2-(3-chlorophenyl)-6-nitro-2H- benzotriazole-1-oxide, 2-(4-chlorophenyl)-6-  
 nitro-2H- benzotriazole-1-oxide, 2-(4-chlorophenyl)-6-picrylazo-2H- benzotriazole-1-oxide, 5-  
 chloro-2-(2,4,5-trimethylphenyl)-2H- benzotriazole-1-oxide, 4,5-dibromo-6-nitro-2-p-tolyl-2H-  
 5 benzotriazole-1-oxide, 4,5-dichloro-6-nitro-2-phenyl-2H- benzotriazole-1-oxide, 4,5-dichloro-6-  
 nitro-2-p-tolyl-2H- benzotriazole-1-oxide, 4,7-dichloro-6-nitro-2-p-tolyl-2H- benzotriazole-1-oxide,  
 4,7-dimethyl-6-nitro-2-phenyl-2H- benzotriazole-1-oxide, 2-(2,4-dimethylphenyl)-4,6- dinitro-2H-  
 benzotriazole-1-oxide, 2-(2,5)-dimethylphenyl)-4,6- dinitro-2H-benzotriazole-1-oxide, 2-(2,4-  
 dimethylphenyl)-6- nitro-2H-benzotriazole-1-oxide, 2-(2,5)-dimethylphenyl)-6-nitro-2H-  
 10 benzotriazole-1-oxide, 4,6-dinitro-2-[3-nitro-4-(N'- phenylhydrazino)phenyl]-2H-benzotriazole-1-  
 oxide, 4,6- dinitro-2-[4-nitro-4-(N'-phenylhydrazino)phenyl]-2H- benzotriazole-1-oxide, 4,6-dinitro-  
 2-phenyl-2H-benzotriazole- 1-oxide, 2-(2,4-dinitrophenyl)-4,6-dinitro-2H-benzotriazole- 1-oxide, 2-  
 (2,4-dinitrophenyl)-6-nitro-2H-benzotriazole-1- oxide, 4,6-dinitro-2-o-tolyl-2H-benzotriazole-1-  
 oxide, 4,6- dinitro-2-p-tolyl-2H-benzotriazole-1-oxide, 4,6-dinitro-2- (2,4,5-trimethylphenyl)-2H-  
 15 benzotriazole-1-oxide, 2-(4- methoxyphenyl)-2H-benzotriazole-1-oxide, 2-(4-methoxyphenyl)- 6-  
 methyl-2H-benzotriazole-1-oxide, 5-methyl-6-nitro-2-m- tolyl-2H-benzotriazole-1-oxide, 5- methyl-  
 6-nitro-2-m-tolyl- 2H-benzotriazole-1-oxide, 5-methyl-6-nitro-2-o-tolyl-2H- benzotriazole-1-oxide,  
 5-methyl-6-nitro-2-p-tolyl-2H- benzotriazole-1-oxide, 6-methyl-4-nitro-2-p-tolyl-2H- benzotriazole-  
 1-oxide, 6-methyl-2-phenyl-2H-benzotriazole-1- oxide, 4-methyl-2-m-tolyl-2H-benzotriazole-1-  
 20 oxide, 4-methyl- 2-o-tolyl-2H-benzotriazole-1-oxide, 4-methyl-2-p-tolyl-2H- benzotriazole-1-oxide,  
 6-methyl-2-m-tolyl-2H-benzotriazole-1- oxide, 6-methyl-2-o-tolyl-2H-benzotriazole-1-oxide, 6-  
 methyl- 2-p-tolyl-2H-benzotriazole-1-oxide, 2-[1]naphthyl-4,6- dinitro-2H-benzotriazole-1-oxide, 2-  
 [2]naphthyl-4,6-dinitro- 2H-benzotriazole-1-oxide, 2-[1]naphthyl-6-nitro-2H- benzotriazole-1-oxide,

2-[2]naphthyl-6-nitro-2H- benzotriazole-1-oxide, 2-(3-nitrophenyl)-2H-benzotriazole-1- oxide, 6-nitro-2-phenyl-2H-benzotriazole-1-oxide, 4-nitro-2- p-tolyl-2H-benzotriazole-1-oxide, 6-nitro-2-o-tolyl-2H- benzotriazole-1-oxide, 6-nitro-2-p-tolyl-2H-benzotriazole-1- oxide, 6-nitro-2-(2,4,5-trimethylphenyl)-2H-benzotriazole-1- oxide, 2-phenyl-2H-benzotriazole-1-oxide, 2-o-tolyl-2H-benzotriazole-1-oxide, 2-p-tolyl-2H-benzotriazole-1-oxide.

Also preferred are condensed N-heterocyclic compounds, such as triazolo and tetrazolo compounds, which can contain at least one N-hydroxy, oxime, N-oxy, N,N-dioxy function and along with N a further hetero atom such as O, S, Se, or Te.

Examples for this are:

[1,2,4]triazolo[4,3-a]pyridine,  
 [1,2,4]triazolo[1,5-a]pyridine,  
 [1,2,4]triazolo[4,3-a]quinoline,  
 [1,2,4]triazolo[4,3-b]isoquinoline,  
 [1,2,4]triazolo[3,4-a]isoquinoline,  
 [1,2,4]triazolo[1,5-b]isoquinoline,  
 [1,2,4]triazolo[5,1-a]isoquinoline,  
 [1,2,3]triazolo[1,5-a]pyridine,  
 [1,2,3]triazolo[4,5-b]pyridine,  
 [1,2,3]triazolo[4,5-c]pyridine,  
 [1,2,3]triazolo[1,5-a]quinoline,  
 [1,2,3]triazolo[5,1-a]isoquinoline,  
 [1,2,4]triazolo[4,3-b]pyridazine,  
 [1,2,4]triazolo[1,5-b]pyridazine,



[1,2,4]triazolo[4,5-d]pyridazine,

[1,2,4]triazolo[4,3-b]quinoline,

[1,2,4]triazolo[3,4-a]phthalazine,

[1,2,4]triazolo[4,3-a]pyrimidine,

5 [1,2,4]triazolo[4,3-c]pyrimidine,

[1,2,4]triazolo[1,5-a]pyrimidine,

[1,2,4]triazolo[1,5-c]pyrimidine,

[1,2,4]triazolo[4,3-c]quinazoline,

[1,2,4]triazolo[1,4-a]quinazoline,

10 [1,2,4]triazolo[1,5-c]quinazoline,

[1,2,4]triazolo[5,1-b]quinazoline,

[1,2,3]triazolo[1,5-a]pyrimidine,

[1,2,3]triazolo[1,5-c]pyrimidine,

[1,2,3]triazolo[4,5-d]pyrimidine,

15 [1,2,3]triazolo[1,5-a]quinazoline,

[1,2,3]triazolo[1,5-c]quinazoline,

[1,2,4]triazolo[4,3-a]pyrazine,

[1,2,4]triazolo[1,5-a]pyrazine,

[1,2,3]triazolo[4,5-b]pyrazine,

20 [1,2,4]triazolo[4,3-a]quinoxaline,

[1,2,3]triazolo[1,5-a]quinoxaline,

[1,2,4]triazolo[4,3-b][1,2,4]triazine,

[1,2,4]triazolo[3,4-c][1,2,4]triazine,

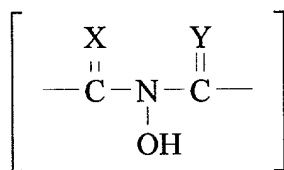
[1,2,4]triazolo[4,3-d][1,2,4]triazine,  
 [1,2,4]triazolo[3,4-f][1,2,4]triazine,  
 [1,2,4]triazolo[1,5-b][1,2,4]triazine,  
 [1,2,4]triazolo[5,1-c][1,2,4]triazine,  
 5 [1,2,4]triazolo[1,5-d][1,2,4]triazine,  
 [1,2,4]triazolo[4,3-a][1,3,5]triazine,  
 [1,2,4]triazolo[1,5-a][1,3,5]triazine,  
 tetrazolo[1,5-a]pyridine,  
 tetrazolo[1,5-b]isoquinoline,  
 10 tetrazolo[1,5-a]quinoline,  
 tetrazolo[5,1-a]isoquinoline,  
 tetrazolo[1,5-b]pyridazine,  
 tetrazolo[1,5-b]quinoline,  
 tetrazolo[5,1-a]phthalazine,  
 15 tetrazolo[1,5-a]pyrimidine,  
 tetrazolo[1,5-c]pyrimidine,  
 tetrazolo[1,5-a]quinazoline,  
 tetrazolo[1,5-c]quinazoline,  
 tetrazolo[1,5-a]pyrazine,  
 20 tetrazolo[1,5-a]quinoxaline,  
 tetrazolo[1,5-b][1,2,4]triazine,  
 tetrazolo[5,1-c][1,2,4]triazine,  
 tetrazolo[1,5-d][1,2,4]triazine,

tetrazolo[5,1-f][1,2,4]triazine.

Other compounds that can be used according to the invention are: quinoline-N-oxide, isoquinoline-N-oxide, N- hydroxy-1,2,3,4-tetrahydroisoquinoline,  $\beta$ -(N-oxy-1,2,3,4-tetrahydroisoquinoline) propionic acid, 1,3-dihydroxy-2N- benzylimidobenzimidazoline.

5 Of the mediators disclosed in International Patent Disclosures WO 94/29425 and DE 4445088.5-43, 1-hydroxy-1H- benzotriazole (HBT) furnishes the best results as a bleach additive in multicomponent systems for use with detergents. However, HBT is available only at high prices and not in adequate amounts. Moreover, with the addition of laccase, for instance, it reacts to form 1H-benzotriazole (BT). This compound is relatively poorly degradable and in major quantities could represent a major environmental burden. Moreover, its reaction speed is not very high and to a certain extent causes damage to the enzymes used. Additionally, HBT causes further colored decomposition products, which are undesirable.

Therefore those mediators that are very particularly preferred (which e.g. exhibit this undesired coloration only to a very limited extent) are those that are selected from the group of cyclical N-hydroxy compounds having at least one optionally substituted 5- or 6-member ring, containing the structure given in formula A:



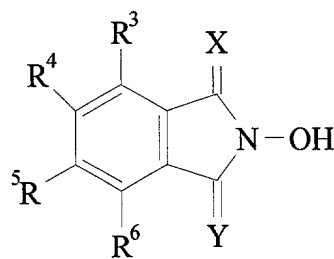
Formula A

as well as their salts, ethers or ester, in which X and Y are the same or different and stand for O, S or NR<sup>1</sup>, in which

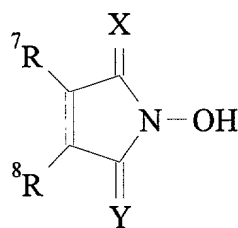
R<sup>1</sup> stands for hydrogen, hydroxyl, formyl, carbamoyl, or sulfono radical, or ester or salt of the sulfono radical, sulfamoyl, nitro, amino, phenyl, aryl C<sub>1</sub>-C<sub>5</sub> alkyl, C<sub>1</sub>-C<sub>12</sub> alkyl, C<sub>1</sub>-C<sub>5</sub> alkoxy, C<sub>1</sub>-C<sub>10</sub> carbonyl, carbonyl C<sub>1</sub>-C<sub>6</sub> alkyl, phospho, phosphono or phosphonooxy radical, or ester or salt of the phosphonooxy radical; and carbamoyl, sulfamoyl, amino and phenyl radicals may be unsubstituted  
 5 or substituted once or multiple times with a radical R<sup>2</sup>, and the aryl C<sub>1</sub>-C<sub>5</sub> alkyl, C<sub>1</sub>-C<sub>12</sub> alkyl, C<sub>1</sub>-C<sub>5</sub> alkoxy, C<sub>1</sub>-C<sub>10</sub> carbonyl, carbonyl C<sub>1</sub>-C<sub>6</sub> alkyl radicals may be saturated or unsaturated, branched or unbranched, and substituted once or multiple times with a radical R<sup>2</sup> and R<sup>2</sup> is the same or different and stands for hydroxyl, formyl, or carboxyl radical, ester or salt of the carboxy radical, carbamoyl, sulfono ester or salt of the sulfono radical, sulfamoyl, nitro, amino, phenyl, C<sub>1</sub>-C<sub>5</sub> alkyl, C<sub>1</sub>-C<sub>5</sub> alkoxy  
 10 radical.

The multicomponent systems of the invention contain mediators that are available on a large industrial scale and are less expensive than HBT. These mediators react under the influence of oxidation agents to form products without any problematic discoloration. These products in turn are completely degradable.

15 The multicomponent system according to the invention includes, as the mediator (component c), at least one compound of the general formula VI, VII, VIII or IX,



VI

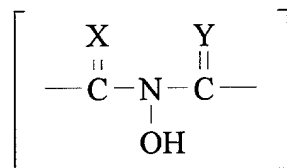


VII



5 and stand for halogen radical, carboxy radical, salt or ester of a carboxy radical, or the meaning given for R<sup>1</sup>; in which R<sup>9</sup> and R<sup>10</sup>, or R<sup>11</sup> and R<sup>12</sup>, must not at the same time stand for a hydroxyl or amino radical and optionally two at a time of the substituents R<sup>3</sup> to R<sup>6</sup>, R<sup>7</sup> to R<sup>8</sup>, R<sup>9</sup> to R<sup>12</sup>, R<sup>13</sup> to R<sup>18</sup> can be linked together into a ring -B-, in which -B- has one of the following meanings:

10



### Formula A

and in which optionally the radicals  $R^9$  to  $R^{12}$  may also be linked to one another by one or two bridge elements -Q-, in which -Q- may be the same or different and can have the following meanings: -O-, -S-,  $CH_2$ -,  $-CR^{19}=CR^{20}$ -, in which  $R^{19}$  and  $R^{20}$  are the same or different and have the same meaning as  $R^3$ .

Compounds that are especially preferred as mediators are those of the general formulas VI, VII, VIII or IX, in which X and Y stand for O or S.

Examples of such compounds are N-hydroxyphthalimide, and optionally substituted N-hydroxyphthalimide derivatives, N-hydroxymaleimide, optionally substituted N-hydroxymaleimide derivatives, N-hydroxynaphthalic acid imide, optionally substituted N-hydroxynaphthalic acid imide derivatives, N-hydroxysuccinimide, optionally substituted N-hydroxysuccinimide derivatives, preferably those in which the radicals  $R^9$  to  $R^{12}$  are polycyclically bonded.

N-Hydroxyphthalimide is especially preferred as the mediator (component c of the multicomponent system of the invention).

Compounds of formula VI that are especially suitable as mediators are for example:

N-hydroxyphthalimide, N-hydroxybenzene-1,2,4-tricarboxylic acid imide, N-N'-dihydroxypyromellitic acid diimide, N-N'-dihydroxybenzophenone-3,3',4,4'-tetracarboxylic acid diimide.

Examples of compounds of formula VII suitable as mediators are:

N-hydroxymaleimide, pyridine-2,3-dicarboxylic acid-N-hydroxyimide.

Examples of compounds of formula VIII suitable as mediators are:

N-hydroxysuccinimide, N-hydroxytartaric acid imide, N-hydroxy-5-norbornene-2,3-dicarboxylic acid imide, exo-N-hydroxy-7-oxabicyclo[2.2.1]-hept-5-ene-2,3-dicarboximide, N-hydroxy-cis-cyclohexane-1,2-dicarboximide, N-hydroxy-cis-4-cyclohexane-1,2-dicarboxylic acid imide.

A compound of formula IX suitable as a mediator is for instance:

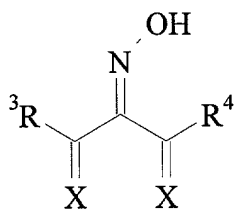
N-hydroxynaphthalic acid imide sodium salt.

A compound with a six-member ring containing the structure given in formula A suitable as a mediator is for instance:

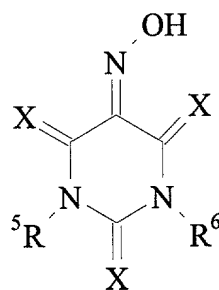
N-hydroxyglutarimide.

The compounds named as examples are also suitable as mediators in the form of their salts or esters.

Mediators that are very particularly preferred, also because of their low cost, good degradability, substantially lower "damage potential" to enzymes, and very fast reaction speed, are those characterized in that they are selected from the group of oximes of the general formula X or XI



X



XI

and their salts, ethers or esters, in which X is the same or different and stands for O, S or NR<sup>1</sup>, in which R<sup>1</sup> stands for hydrogen, hydroxyl, formyl, carbamoyl, or sulfono radical, or ester or salt of the sulfono radical, sulfamoyl, nitro, amino, phenyl, acryl C<sub>1</sub>-C<sub>5</sub> alkyl, C<sub>1</sub>-C<sub>12</sub> alkyl, C<sub>1</sub>-C<sub>5</sub> alkoxy, C<sub>1</sub>-C<sub>10</sub> carbonyl, carbonyl C<sub>1</sub>-C<sub>6</sub> alkyl, phospho, phosphono or phosphonooxy radical, or ester or salt of the phosphonooxy radical; in which carbamoyl, sulfamoyl, amino and phenyl radicals may be unsubstituted or substituted once or multiple times with a radical R<sup>2</sup>, and the aryl C<sub>1</sub>-C<sub>5</sub> alkyl, C<sub>1</sub>-C<sub>12</sub> alkyl, C<sub>1</sub>-C<sub>5</sub> alkoxy, C<sub>1</sub>-C<sub>10</sub> carbonyl, carbonyl C<sub>1</sub>-C<sub>6</sub> alkyl radicals may be saturated or unsaturated, branched or unbranched, and substituted once or multiple times with a radical R<sup>2</sup>; and

R<sup>2</sup> is the same or different and stands for hydroxy, formyl, or carboxy radical, ester or salt of the carboxy radical, carbamoyl, sulfono ester or salt of the sulfono radical, sulfamoyl, nitro, amino, phenyl, C<sub>1</sub>-C<sub>5</sub> alkyl, C<sub>1</sub>-C<sub>5</sub> alkoxy radical, and the radicals R<sup>3</sup> and R<sup>4</sup> are the same or different and

stand for halogen, carboxyl radical, ester or salt of the carboxyl radical, or have the meanings given for  $R^1$ , or are linked together into a ring  $(-CR^7R^8)_n$ , where n is equal to 2, 3 or 4, and

$R^5$  and  $R^6$  have the meanings given for  $R_1$ , and

$R^7$  and  $R^8$  are the same or different and stand for halogen, carboxyl radical, ester or salt of the carboxyl radical, or have the meanings given for  $R^1$ .

Especially preferred as mediators in the multicomponent system of the invention are compounds with the general formula X, in which X stands for O or S and the other radicals have the meanings named above. One example of such a compound is 2-hydroxyiminomalonic acid dimethyl ester.

Also especially preferred as mediators are isonitro derivatives of cyclical ureides of general formula XI. Examples of such compounds are 1-methylvioluric acid, 1,3- dimethylvioluric acid, thiovioluric acid, and alloxane-4-5- dioxime.

Especially preferred as a mediator is alloxane-5-oxime hydrate (violuric acid) and/or its esters or salts.

#### Comediators

The components d) can contain for instance aliphatic ethers and/or aryl-substituted alcohols, such as:

2,3- dimethoxybenzyl alcohol, 3,4-dimethoxybenzyl alcohol, 2,4- dimethoxybenzyl alcohol, 2,6-dimethoxybenzyl alcohol, homovanillyl alcohol, ethylene glycol monophenyl ether, 2-hydroxybenzyl alcohol, 4-hydroxybenzyl alcohol, 4-hydroxy-3- methoxybenzyl alcohol, 2-methoxybenzyl alcohol, 2,5- dimethoxybenzyl alcohol, 2,4-dimethoxybenzylamine, 2,4-dimethoxybenzylamine hydrochloride, veratryl alcohol, and coniferyl alcohol.



Olefins (alkenes) can also be considered, such as 2- allylphenol, 2-allyl-6-methylphenol, allylbenzene, 3,4- dimethoxypropanylbenzene, p-methoxystyrene, 1-allylimidazol, 1-vinylimidazol, styrene, stilbene, allylphenyl ether, cinnamic acid benzyl ester, cinnamic acid methyl ester, 2,4,6-triallyloxy-1,3,5-triazine, 1,2,4-trivinylcyclohexane, 4-allyl-1,2-dimethoxybenzene, 4-tert-butylbenzoic acid vinyl ester, squalene, benzoin allyl ether, cyclohexene, dihydropyran, and N-benzylcinnamic acid anilide.

Phenol ethers are preferably used, such as 2,3- dimethoxybenzyl alcohol, 3,4-dimethoxybenzyl alcohol, 2,4- dimethoxybenzyl alcohol, 2,6-dimethoxybenzyl alcohol, homovanillyl alcohol, 4-hydroxybenzyl alcohol, 4-hydroxy-3- methoxybenzyl alcohol, 2-methoxybenzyl alcohol, 2,5- dimethoxybenzyl alcohol, 2,4-dimethoxybenzylamine, 2,4-dimethoxybenzylamine hydrochloride, veratryl alcohol, coniferyl alcohol, veratrol, and anisol.

Also preferred are carbonyl compounds, such as 4- aminobenzophenone, 4-acetylbiphenyl, benzophenone, benzil, benzophenone hydrazone, 3,4-dimethoxybenzaldehyde, 3,4-dimethoxybenzoic acid, 3,4-dimethoxybenzophenone, 4- dimethylaminobenzaldehyde, 4-acetylbiphenylhydrazone, benzophenone-4-carboxylic acid, benzoyl acetone, bis-(4,4-dimethylamino)benzophenone, benzoin, benzoin oxime, N- benzoyl-N-phenylhydroxyl-amine, 2-amino-5-chlorobenzophenone, 3-hydroxy-4-methoxybenzaldehyde, 4-methoxybenzaldehyde, anthraquinone-2-sulfonic acid, 4-methylaminobenzaldehyde, benzaldehyde, benzophenone-2-carboxylic acid, 3,3,'4,4'- benzophenonetetracarboxylic acid dianhydride, (S)-(-)-2-(N-benzylpropyl)-aminobenzophenone, benzylphenyl acetic acid anilide, N-benzylbenzanilide, 4,4'-bis-(dimethylamino)thiobenzophenone, 4,4- bis(diacetylamino)benzophenone, 2-chlorobenzophenone, 4,4'- dihydroxybenzophenone, 2,4-dihydroxybenzophenone, 3,5- dimethoxy-4-hydroxybenzaldehyde hydrazine, 4- hydroxybenzophenone, 4-methoxybenzophenone, 3,4-

dihydroxybenzophenone, p-anisic acid, p-anisic aldehyde, 3,4- dihydroxybenzaldehyde, 3,4- dihydroxybenzoic acid, 3,5- dimethoxy-4-hydroxybenzaldehyde, 3,5-dimethoxy-4- hydroxybenzoic acid, 4-hydroxybenzaldehyde, salicylaldehyde, vanillin, and vannilic acid.

#### Component e)

5 By the addition of the compounds of the multicomponent system that are named under categories d) and e), a reaction mediation in cascade form, or recycling of the actual mediator compounds in situ, that is, during the reaction, ensues, and surprisingly leads to a substantial improvement in the bleaching reaction.

10 The multicomponent system contains as a free amine, in the case of the in situ generation or reaction mediation in cascade form for hydroxybenzotriazol benzotriazole.

#### Further Components

In addition, the bleaching system may contain phenolic compounds and/or nonphenolic compounds with one or more benzene nuclei.

15 Along with the oxidation agents named above according to the invention, air, oxygen,  $H_2O_2$ , organic peroxides, sodium perborate, and/or sodium percarbonate are especially preferred.

Oxygen can also be generated in situ by means of  $H_2O_2$ + catalase or similar systems or  $H_2O_2$  from GOD + glucose or similar systems.

Also preferred is a multicomponent bleaching system containing cation-forming metal salts. As cations,  $Fe^{2+}$ ,  $Fe^{3+}$ ,  $Mn^{2+}$ ,  $Mn^{3+}$ ,  $Mn^{4+}$ ,  $Cu^+$ ,  $Cu^{2+}$ ,  $Ti^{3+}$ ,  $Cer^{4+}$ ,  $Mg^{2+}$ , and  $Al^{3+}$  should be used.

20 The bleaching system can additionally contain polysaccharides and/or proteins. As polysaccharides, it is possible to use, among others, glucanes, mannanes, dextrans, levans, pectins, alginates, or vegetable rubbers, and/or its own polysaccharides formed by the fungi or produced in a mixed culture with yeasts. As proteins, gelatin or albumin among others can be used.

Other possible substances are simple sugar, oligomer sugar, amino acids, polyethylene glycoles (PEG's), polyethylene oxides, polyethylene imines and polydimethyl siloxanes.

#### Use of the Multicomponent System

5 The multicomponent bleaching system of the invention can also be used in combination with detergent additives known per se.

The bleaching system develops its effect in a pH range from 2 to 12, preferably from 4 to 10, and at a temperature between 10°C and 60°C, and preferably between 20°C and 40°C.

The invention will be described in further detail below with reference to the examples:

#### Example 1

10 Influence of the laccase mediator system on standard cotton fabrics stained with coffee.

Example: In 100 ml of washing solution (in a 300 ml Erlenmeyer flask), one fabric cloth each (5 x 5 cm) is incubated at 40°C for 40 minutes while being shaken with reciprocation (120 rpm). Before the onset of incubation, the washing solution is subjected to a ten-minute temperature adaptation. The washing solution is made up with STW (standard tap water) at 14° dH ("dH" means 15 German degree of hardness). As the enzyme dosage, 200 IU\* of laccase from Coriolus versicolor per 100 ml is used, and as the mediator dosage, 200 mg of hydroxybenzotriazole per 100 ml is used.

After the "washwater" is poured off, the flask is filled with a cold, strong stream of water three times and poured off again.

20 Table 1 shows the results, in comparison with a commercial liquid detergent (without a bleaching system) and a solid detergent (with a bleaching system).

\* 1 IU = conversion of 1  $\mu$  mol of Syringaldazine per min per ml enzyme

Table 1

	pH	Whiteness	Brightness
STW zero value	4.5	2.55	2.3
Solid detergent	10.1	8.9	6.15
STW + enzyme + mediator	4.5	5.0	5.8
Liquid detergent	4.5	3.85	3.75
Liquid detergent + enzyme + mediator	4.5	6.15	6.6

### Example 2

Influence of the laccase mediator system on standard tea fabrics stained with coffee.

In 100 ml of washing solution (in a 300 ml Erlenmeyer flask), one fabric cloth each (5 x 5  
5 cm) is incubated at 40°C for 40 minutes while being shaken with reciprocation (120 rpm).

Before the onset of incubation, the washing solution is subjected to a ten-minute temperature adaptation. The washing solution is made up with STW (standard tap water) at 14° dH. As the enzyme dosage, 200 IU of laccase from *Coriolus versicolor* per 100 ml is used, and as the mediator dosage, 200 mg of hydroxybenzotriazole per 100 ml is used.

After the “washwater” is poured off, the flask is filled with a cold, strong stream of water  
three times and poured off again.

Table 2 shows the results, in comparison with a commercial liquid detergent (without a bleaching system) and a solid detergent (with a bleaching system).

Table 2

	pH	Whiteness	Brightness
STW zero value	4.5	2.7	2.5
Solid detergent	10.1	8.95	8.6
STW + enzyme + mediator	4.5	4.2	4.7
Liquid detergent	4.5	4.7	4.7
Liquid detergent + enzyme + mediator	4.5	5.5	5.95

### Example 3

An experiment was performed in accordance with Example 1.

Acetoxybenzotriazole was used as the mediator.

The results can be found in Table 3.

Table 3

	pH	Whiteness	Brightness
STW zero value	4.5	2.55	2.3
Solid detergent	10.1	8.9	6.15
STW + enzyme + mediator	4.5	5	6.1
Liquid detergent	4.5	3.85	3.75
Liquid detergent + enzyme + mediator	4.5	6.2	6.7

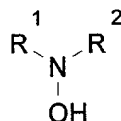
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CLAIMS

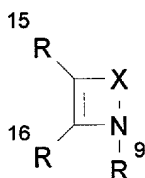
1. A multicomponent system for use with detergents, containing
  - a) optionally, at least one oxidation catalyst;
  - b) at least one suitable oxidizing agent;
  - c) at least one mediator, selected from the group comprising hydroxylamines, hydroxylamine derivatives, hydroxamic acids, hydroxamic acid derivatives, and the aliphatic, cycloaliphatic, heterocyclic or aromatic compounds that contain at least one N-hydroxy, oxime, N-oxy, or N,N'-dioxy function;
  - d) at least one comediator, selected from the group comprising aryl-substituted alcohols, carbonyl compounds, aliphatic ethers, phenol ethers, and/or olefins (alkenes); and
  - e) optionally, a small quantity of at least one free amine of a respective mediator used.
2. The multicomponent system of claim 1, characterized in that in addition to the these substances, it contains phenolic and/or nonphenolic compounds with one or more benzene nuclei.
3. The multicomponent system of claim 1 or 2, characterized in that as the oxidation catalyst, it contains one or more oxidoreductases of classes 1.1.1 - 1.97.
4. The multicomponent system of claim 3, characterized in that it contains one or more oxidoreductases which use oxygen, peroxides or quinones as electron acceptors.
5. The multicomponent system of claim 3, characterized in that as the oxidoreductase, it contains laccase (1.10.3.2.).
6. The multicomponent system of claim 1 or 2, characterized in that component c), as the NO-, NOH- or H-NR- OH-containing aliphatic, cycloaliphatic, heterocyclic or aromatic compounds, it contains N-hydroxy, oxime, N-oxy and N,N'-dioxy compounds in single- or multicomponent systems.

7. The multicomponent system of claim 6, characterized in that component c), as NO-, NOH- or H-NR-OH- containing compounds, contains hydroxylamines of the general formula

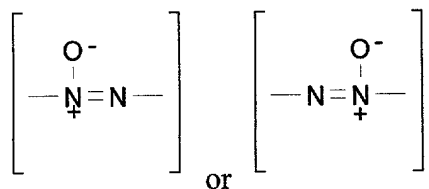


in which the substituents R<sup>1</sup> and R<sup>2</sup>, which may be the same or different, independently of one other represent one of the following groups: hydrogen, C<sub>1</sub>-C<sub>12</sub> alkyl, carbonyl C<sub>1</sub>-C<sub>6</sub> alkyl, phenyl, aryl, of which C<sub>1</sub>-C<sub>12</sub> alkyl, carbonyl C<sub>1</sub>-C<sub>6</sub> alkyl, phenyl, aryl may be unsubstituted or may also be substituted once or multiple times with the radical R<sup>3</sup>, in which the radical R<sup>3</sup> may represent one of the following groups: hydrogen, halogen, hydroxyl, formyl, carboxyl and salts and esters thereof, amino, nitro, C<sub>1</sub>-C<sub>12</sub> alkyl, C<sub>1</sub>-C<sub>6</sub> alkyloxy, carbonyl C<sub>1</sub>-C<sub>6</sub> alkyl, phenyl, sulfono, their esters and salts, sulfamoyl, carbamoyl, phospho, phosphono, phosphonooxy and their salts and esters; in which the amino, carbamoyl and sulfamoyl groups of the radical R<sup>3</sup> may be unsubstituted or may be substituted once or two times with hydroxyl, C<sub>1</sub>-C<sub>3</sub> alkyl, C<sub>1</sub>-C<sub>3</sub> alkoxy; in which the radicals R<sup>1</sup> and R<sup>2</sup> can jointly form a group-B-, and -B- in that case represents one of the following groups: (-CHR<sup>4</sup>-)<sub>n</sub>, (CR<sup>4</sup>=CH-)<sub>m</sub>; and in which R<sup>4</sup> is a substituent that is defined line R<sup>3</sup>, and n represents an integer from 1 to 6 and m represents an integer from 1 to 3.

8. The multicomponent system of claim 6, characterized in that component c), as NO-, NOH- or H-NR-OH- containing compounds, contains substances of the general formula



in which X stands for one of the following groups: (-N=N-), (-N=CR<sup>10</sup>-)<sub>p</sub>, (-CR<sup>10</sup>=N-)<sub>p</sub>, (-CR<sup>11</sup>=CR<sup>12</sup>-)<sub>p</sub>



and p is equal to 1 or 2, in which the radicals R<sup>9</sup> to R<sup>12</sup>, R<sup>15</sup> and R<sup>16</sup> may be the same or different and

10 independently of one another can represent one of the following groups: hydrogen, halogen, hydroxyl, formyl, carboxyl and salts and esters thereof; amino, nitro, C<sub>1</sub>-C<sub>12</sub> alkyl, C<sub>1</sub>-C<sub>6</sub> alkyloxy, carbonyl C<sub>1</sub>-C<sub>6</sub> alkyl, phenyl, sulfono esters and salts thereof, sulfamoyl, carbamoyl, phospho, phosphono, phosphonooxy and their salts and esters; and in which the amino, carbamoyl and sulfamoyl groups of the radicals R<sup>9</sup> to R<sup>12</sup>, R<sup>15</sup> and R<sup>16</sup> may be unsubstituted or may also be

15 substituted once or two times with hydroxyl, C<sub>1</sub>-C<sub>3</sub> alkyl, C<sub>1</sub>-C<sub>3</sub> alkoxy; and in which the radicals R<sup>15</sup> and R<sup>16</sup> can form a common group -G-, and -G- represents one of the following groups: (-CR<sup>5</sup>=CR<sup>6</sup>-CR<sup>7</sup>=CR<sup>8</sup>-) or (-CR<sup>8</sup>=CR<sup>7</sup>-CR<sup>6</sup>=CR<sup>5</sup>-), in which the radicals R<sup>5</sup> to R<sup>8</sup> may be the same or different and independently of one another can represent one of the following groups: hydrogen, halogen, hydroxyl, formyl, carboxyl and salts and esters thereof; amino, nitro, C<sub>1</sub>-C<sub>12</sub> alkyl, C<sub>1</sub>-C<sub>6</sub> alkyloxy, carbonyl C<sub>1</sub>-C<sub>6</sub> alkyl, phenyl, sulfono, esters and salts thereof, sulfamoyl, carbamoyl, phospho,

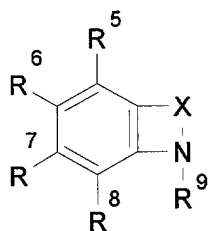
20 phosphono, phosphonooxy and their salts and esters, and in which the amino, carbamoyl and sulfamoyl groups of the radicals R<sup>5</sup> to R<sup>8</sup> may be unsubstituted or may also be substituted once or two times with hydroxyl, C<sub>1</sub>-C<sub>3</sub> alkyl, C<sub>1</sub>-C<sub>3</sub> alkoxy; and in which the C<sub>1</sub>-C<sub>12</sub> alkyl, C<sub>1</sub>-C<sub>6</sub> alkyloxy, carbonyl C<sub>1</sub>-C<sub>6</sub> alkyl, phenyl, aryl groups of radicals R<sup>5</sup> to R<sup>8</sup> may be unsubstituted or may also be

25 substituted one or two times with the radical R<sup>18</sup>; in which the radical R<sup>18</sup> can represent one of the following groups: hydrogen, halogen, hydroxyl, formyl, carboxyl and their salts and esters; amino, nitro, C<sub>1</sub>-C<sub>12</sub> alkyl, C<sub>1</sub>-C<sub>6</sub> alkyloxy, carbonyl C<sub>1</sub>-C<sub>6</sub> alkyl, phenyl, aryl, and their esters and salts, and

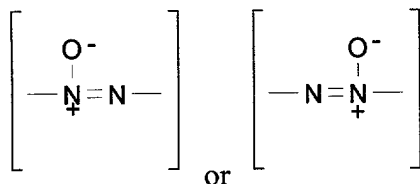


the carbamoyl, sulfamoyl, amino groups of the radical  $R^{18}$  may be unsubstituted or may also be substituted once or two times with the radical  $R^{19}$  and the radical  $R^{19}$  may represent one of the following groups: hydrogen; hydroxyl, formyl, carboxyl and their salts and esters; amino, nitro,  $C_1$ - $C_{12}$  alkyl,  $C_1$ - $C_6$  alkyloxy, carbonyl  $C_1$ - $C_6$  alkyl, phenyl, aryl.

9. The multicomponent system of claim 6, characterized in that the component c), as  $NO$ -,  $NOH$ - or  $H-NR$ -  $OH$ -containing compounds, contains compounds of the general formula III,



5 in which X stands for one of the following groups:  $(-N=N-)$ ,  $(-N=CR^{10}-)_p$ ,  $(-CR^{10}=N-)_p$ ,  $(-CR^{11}=CR^{12}-)_p$



and p is equal to 1 or 2, in which the radicals  $R^1$  to  $R^{12}$  are same or different and independently of one another can represent one of the following groups: hydrogen, halogen, hydroxyl, formyl,

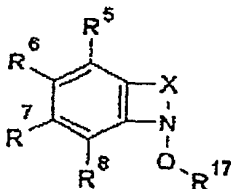
10 carboxyl and salts and esters thereof; amino, nitro,  $C_1$ - $C_{12}$  alkyl,  $C_1$ - $C_6$  alkyloxy, carbonyl  $C_1$ - $C_6$  alkyl, phenyl, aryl, sulfono, esters and salts thereof, sulfamoyl, carbamoyl, phospho, phosphono, phosphonooxy and their salts and esters; and in which their amino, carbamoyl and sulfamoyl groups may be unsubstituted or may also be substituted once or two times with hydroxy,  $C_1$ - $C_3$  alkyl,  $C_1$ - $C_3$  alkoxy; and in which the  $C_1$ - $C_{12}$  alkyl,  $C_1$ - $C_6$  alkyloxy, carbonyl  $C_1$ - $C_6$  alkyl, phenyl, aryl, aryl  $C_1$ - $C_6$  alkyl groups of radicals  $R^5$  to  $R^{12}$  may be unsubstituted or substituted once or two times with the

radical  $R^{13}$ , and the radical  $R^{13}$  can represent one of the following groups: hydrogen, halogen, hydroxyl, formyl, carboxyl and their salts and esters; amino, nitro,  $C_1$ - $C_{12}$  alkyl,  $C_1$ - $C_6$  alkyloxy, carbonyl  $C_1$ - $C_6$  amino, nitro,  $C_1$ - $C_{12}$  alkyl,  $C_1$ - $C_6$  alkyloxy, carbonyl  $C_1$ - $C_6$  alkyl, phenyl, aryl, sulfono, sulfeno, sulfino, and their esters and salts. The carbamoyl, sulfamoyl, amino groups of the

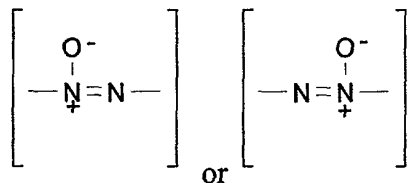
20 radical  $R^{13}$  may be unsubstituted or may also be substituted once or two times with the radical  $R^{14}$ .

The radical  $R^{14}$  may represent one of the following groups: hydrogen; hydroxyl, formyl, carboxyl and their salts and esters; amino, nitro,  $C_1$ - $C_{12}$  alkyl,  $C_1$ - $C_6$  alkyloxy, carbonyl  $C_1$ - $C_6$  alkyl, phenyl or aryl.

10. The multicomponent system of claim 6, characterized in that the component c), as NO-, NOH- or H-NR- OH-containing compounds, contains compounds of the general formula IV,

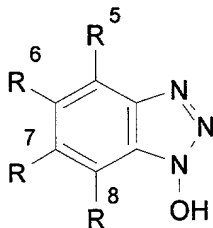


in which X stands for one of the following groups:  $(-N=N-)$ ,  $(-N=CR^{10}-)_p$ ,  $(-CR^{10}=N-)_p$ ,  $(-CR^{11}=CR^{12}-)_p$



10 and p is equal to 1 or 2, in which for the radicals  $R^5$  to  $R^8$  and  $R^{10}$  to  $R^{12}$  the same as in claim 9 applies, and  $R^{17}$  can be hydrogen,  $C_1$ - $C_{10}$  alkyl,  $C_1$ - $C_{10}$  carbonyl, of which  $C_1$ - $C_{10}$  alkyl and  $C_1$ - $C_{10}$  carbonyl can be unsubstituted or mono- or polysubstituted with a radical  $R^{18}$ , which is defined like  $R^3$ .

11. The multicomponent system of claim 6, characterized in that component c), as NO-, NOH- or H-NR-OH- containing compounds, contains 1-hydroxybenzotriazol and the tautomeric benzotriazole-1-oxide, as well as their esters and salts, of the formula



5 in which the radicals  $R^1$  to  $R^8$  may be the same or different and independently of one another can represent one of the following groups: hydrogen, halogen, hydroxyl, formyl, carboxyl and salts and esters thereof; amino, nitro,  $C_1$ - $C_{12}$  alkyl,  $C_1$ - $C_6$  alkyloxy, carbonyl  $C_1$ - $C_6$  alkyl, phenyl, sulfono esters and salts thereof, sulfamoyl, carbamoyl, phospho, phosphono, phosphonooxy and their salts and esters; and in which the amino, carbamoyl and sulfamoyl groups of the radicals  $R^5$  to  $R^8$  may be 10 unsubstituted or may also be substituted once or two times with hydroxyl,  $C_1$ - $C_3$  alkyl,  $C_1$ - $C_3$  alkoxy; and in which the  $C_1$ - $C_{12}$  alkyl,  $C_1$ - $C_6$  alkyloxy, carbonyl  $C_1$ - $C_6$  alkyl, phenyl, aryl groups of radicals  $R^5$  to  $R^8$  may be unsubstituted or may also be substituted or mono- or polysubstituted with the radical  $R^{18}$ , in which the radical  $R^{18}$  can represent one of the following groups: hydrogen, 15 halogen, hydroxyl, formyl, carboxyl and their salts and esters; amino, nitro,  $C_1$ - $C_{12}$  alkyl,  $C_1$ - $C_6$  alkyloxy, carbonyl  $C_1$ - $C_6$  alkyl, phenyl, aryl, sulfono, sulfeno, sulfino, and their esters and salts, and the carbamoyl, sulfamoyl, amino groups of the radical  $R^{18}$  may be unsubstituted or may also be substituted once or two times with the radical  $R^{19}$ , and the radical  $R^{19}$  may represent one of the following groups: hydrogen, hydroxyl, formyl, carboxyl and their salts and esters; amino, nitro,  $C_1$ - 20  $C_{12}$  alkyl,  $C_1$ - $C_6$  alkyloxy, carbonyl  $C_1$ - $C_6$  alkyl, phenyl, aryl.

12. The multicomponent system of claim 6, characterized in that component c), as NO-, NOH- or H-NR-OH- containing compounds, contains such compounds of azoles.

13. The multicomponent system of claim 6, characterized in that component c), as NO-, NOH- or H-NR-OH- containing compounds, contains such compounds of condensed heterocyclic compounds which contain a triazolo or tetrazolo unit, such as:

- 5 [1,2,4]triazolo[4,3-a]pyridine,  
[1,2,4]triazolo[1,5-a]pyridine,  
[1,2,4]triazolo[4,3-a]quinoline,  
[1,2,4]triazolo[4,3-b]isoquinoline,  
[1,2,4]triazolo[3,4-a]isoquinoline,  
10 [1,2,4]triazolo[1,5-b]isoquinoline,  
[1,2,4]triazolo[5,1-a]isoquinoline,  
[1,2,3]triazolo[1,5-a]pyridine,  
[1,2,3]triazolo[4,5-b]pyridine,  
[1,2,3]triazolo[4,5-c]pyridine,  
15 [1,2,3]triazolo[1,5-a]quinoline,  
[1,2,3]triazolo[5,1-a]isoquinoline,  
[1,2,4]triazolo[4,3-b]pyridazine,  
[1,2,4]triazolo[1,5-b]pyridazine,  
[1,2,4]triazolo[4,5-d]pyridazine,  
20 [1,2,4]triazolo[4,3-b]quinoline,  
[1,2,4]triazolo[3,4-a]phthalazine,  
[1,2,4]triazolo[4,3-a]pyrimidine,  
[1,2,4]triazolo[4,3-c]pyrimidine,  
[1,2,4]triazolo[1,5-a]pyrimidine,

25 [1,2,4]triazolo[1,5-c]pyrimidine,  
 [1,2,4]triazolo[4,3-c]quinazoline,  
 [1,2,4]triazolo[1,4-a]quinazoline,  
 [1,2,4]triazolo[1,5-c]quinazoline,  
 [1,2,4]triazolo[5,1-b]quinazoline,  
 30 [1,2,3]triazolo[1,5-a]pyrimidine,  
 [1,2,3]triazolo[1,5-c]pyrimidine,  
 [1,2,3]triazolo[4,5-d]pyrimidine,  
 [1,2,3]triazolo[1,5-a]quinazoline,  
 [1,2,3]triazolo[1,5-c]quinazoline,  
 35 [1,2,4]triazolo[4,3-a]pyrazine,  
 [1,2,4]triazolo[1,5-a]pyrazine,  
 [1,2,3]triazolo[4,5-b]pyrazine,  
 [1,2,4]triazolo[4,3-a]quinoxaline,  
 [1,2,3]triazolo[1,5-a]quinoxaline,  
 40 [1,2,4]triazolo[4,3-b][1,2,4]triazine,  
 [1,2,4]triazolo[3,4-c][1,2,4]triazine,  
 [1,2,4]triazolo[4,3-d][1,2,4]triazine,  
 [1,2,4]triazolo[3,4-f][1,2,4]triazine,  
 [1,2,4]triazolo[1,5-b][1,2,4]triazine,  
 45 [1,2,4]triazolo[5,1-c][1,2,4]triazine,  
 [1,2,4]triazolo[1,5-d][1,2,4]triazine,  
 [1,2,4]triazolo[4,3-a][1,3,5]triazine,

50

tetrazolo[1,5-b]isoquinoline,

tetrazolo[5,1-a]isoquinoline,

tetrazolo[1,5-b]quinoline,

tetrazolo[5,1-a]phthalazine,

tetrazolo[1,5-c]pyrimidine,

tetrazolo[1,5-c]quinazoline,

tetrazolo[1,5-a]pyrazine,

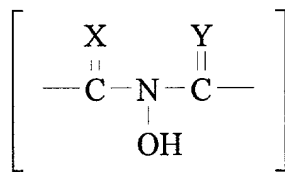
tetrazolo[1,5-b][1,2,4]triazine,

tetrazolo[5,1-c][1,2,4]triazine,

tetrazolo[1,5-d][1,2,4]triazine,

tetrazolo[5,1-f][1,2,4]triazine.

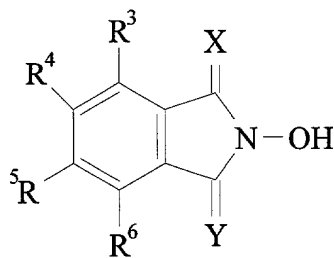
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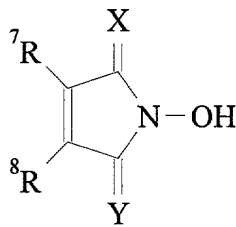
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as well as their salts, ethers or ester, in which X and Y are the same or different and stand for O, S or NR<sup>1</sup>, in which R<sup>1</sup> stands for hydrogen, hydroxyl, formyl, carbamoyl, or sulfono radical, or ester or salt of the sulfono radical, sulfamoyl, nitro, amino, phenyl, aryl C<sub>1</sub>-C<sub>5</sub> alkyl, C<sub>1</sub>-C<sub>12</sub> alkyl, C<sub>1</sub>-C<sub>5</sub> alkoxy, C<sub>1</sub>-C<sub>10</sub> carbonyl, carbonyl C<sub>1</sub>-C<sub>6</sub> alkyl, phospho, phosphono or phosphonooxy radical, or ester or salt of the phosphonooxy radical; in which carbamoyl, sulfamoyl, amino and phenyl radicals may be unsubstituted or substituted once or multiple times with a radical R<sup>2</sup>, and the aryl C<sub>1</sub>-C<sub>5</sub> alkyl, C<sub>1</sub>-C<sub>12</sub> alkyl, C<sub>1</sub>-C<sub>5</sub> alkoxy, C<sub>1</sub>-C<sub>10</sub> carbonyl, carbonyl C<sub>1</sub>-C<sub>6</sub> alkyl radicals may be saturated or unsaturated, branched or unbranched, and substituted once or multiple times with a radical R<sup>2</sup>, and R<sup>2</sup> is the same or different and stands for hydroxyl, formyl, or carboxyl radical, ester or salt of the carboxyl radical, carbamoyl, sulfono ester or salt of the sulfono radical, sulfamoyl, nitro, amino, phenyl, C<sub>1</sub>-C<sub>5</sub> alkyl, C<sub>1</sub>-C<sub>5</sub> alkoxy radical.

15. The multicomponent system of claim 6 or 14, characterized in that as the mediator (component c), at least one compound of the general formula VI, VII, VIII or IX is used:

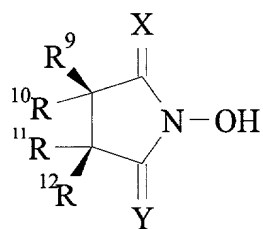


VI

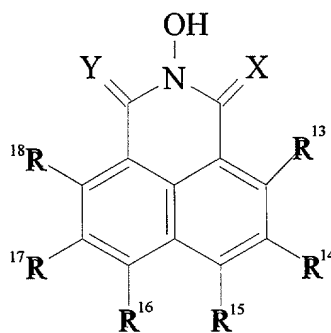


VII

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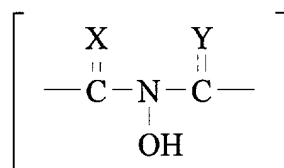
VIII



IX

in which X, Y have the meanings already given and the radicals  $R^3$  to  $R^{18}$  are the same or different and stand for halogen radical, carboxyl radical, salt or ester of a carboxyl radical, or the meaning given for  $R^1$ ; in which  $R^9$  and  $R^{10}$ , or  $R^{11}$  and  $R^{12}$ , must not at the same time stand for a hydroxyl or amino radical, and optionally two at a time of the substituents  $R^3$  to  $R^6$ ,  $R^7$  to  $R^8$ ,  $R^9$  to  $R^{12}$ ,  $R^{13}$  to  $R^{18}$  can be linked together into a ring -B-, in which -B- has one of the following meanings:

$(-CH=CH)_n$ , where  $n = 1-3$ ,  $-CH=CH-CH=N-$ , or



Formula A and in which optionally the radicals  $R^9$  to  $R^{12}$  may also be linked to one another by one or two bridge elements -Q-, in which -Q- may be the same or different and can have the following meanings: -O-, -S-,  $\text{CH}_2$ -,  $-\text{CR}^{19}=\text{CR}^{20}-$ ; in which  $R^{19}$  and  $R^{20}$  are the same or different and have the same meaning as  $R^3$ .

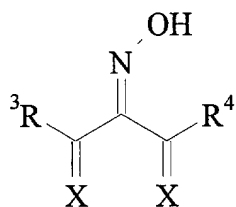
16. The multicomponent system of claim 6, 14, 15, characterized in that as the mediator, at least one substance, selected from the group comprising N- hydroxyphthalimide, optionally substituted N- hydroxyphthalimide derivatives, N-hydroxymaleimide, optionally substituted N-



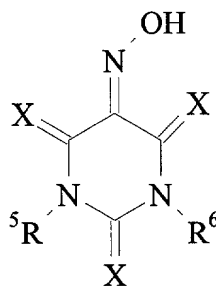
hydroxymaleimide derivatives, N- hydroxynaphthalic acid imide, optionally substituted N-

- 5 hydroxynaphthalic acid imide derivatives, N- hydroxysuccinimide, optionally substituted N-  
hydroxysuccinimide derivatives, is used.

17. The multicomponent bleaching system of claim 6, characterized in that as the  
mediators (component c), oximes of the general formula X or XI



X



XI

and their salts, ethers or esters are used, in which X is the same or different and stands for O, S or  
NR<sup>1</sup>, in which R<sup>1</sup> stands for hydrogen, hydroxyl, formyl, carbamoyl, or sulfono radical, or ester or  
salt of the sulfono radical, sulfamoyl, nitro, amino, phenyl, aryl C<sub>1</sub>-C<sub>5</sub> alkyl, C<sub>1</sub>-C<sub>12</sub> alkyl, C<sub>1</sub>-C<sub>5</sub>  
alkoxy, C<sub>1</sub>-C<sub>10</sub> carbonyl, carbonyl C<sub>1</sub>-C<sub>6</sub> alkyl, phospho, phosphono or phosphonooxy radical, or  
ester or salt of the phosphonooxy radical, in which carbamoyl, sulfamoyl, amino and phenyl radicals  
may be unsubstituted or substituted once or multiple times with a radical R<sup>2</sup>, and the aryl C<sub>1</sub>-C<sub>5</sub> alkyl,  
C<sub>1</sub>-C<sub>12</sub> alkyl, C<sub>1</sub>-C<sub>5</sub> alkoxy, C<sub>1</sub>-C<sub>10</sub> carbonyl, carbonyl C<sub>1</sub>-C<sub>6</sub> alkyl radicals may be saturated or  
unsaturated, branched or unbranched, and substituted once or multiple times with a radical  
R<sup>2</sup>, and R<sup>2</sup> is the same or different and stands for hydroxyl, formyl, or carboxyl radical, ester or salt  
of the carboxy radical, carbamoyl, sulfono ester or salt of the sulfono radical, sulfamoyl, nitro,  
amino, phenyl, C<sub>1</sub>-C<sub>5</sub> alkyl, C<sub>1</sub>-C<sub>5</sub> alkoxy radical, and the radicals R<sup>3</sup> and R<sup>4</sup> are the same or different

and stand for halogen, carboxyl radical, ester or salt of the carboxyl radical, or have the meanings given for  $R^1$ , or are linked together into a ring  $(-CR^7R^8)_n$ , where  $n$  is equal to 2, 3 or 4, and

$R^5$  and  $R^6$  have the meanings given for  $R_1$ , and  $R^7$  and  $R^8$  are the same or different and stand for

20 halogen, carboxyl radical, ester or salt of the carboxyl radical, or have the meanings given for  $R^1$ .

18. The multicomponent bleaching system of one of claims 6 or 17, characterized in that as the mediator, compounds of the general formula X, in which X stands for O or S, and the other radicals have the above-given meanings, are used.

19. The multicomponent system of one of claims 6, 17 or 18, characterized in that as the mediator, isonitroso derivatives of cyclical ureides of the general formula XI are used.

20. The multicomponent bleaching system of one of claims 6, 17 to 19, characterized in that as the mediator, alloxane-5-oxime hydrate (violuric acid) or its esters or salts are used.

21. The multicomponent system of claim 1 or 2, characterized in that as the oxidizing agent, it contains for instance air, oxygen, ozone,  $H_2O_2$ , organic peroxides, peracids such as peracetic acid, performic acid, persulfuric acid, pernitric acid, metachloroperoxybenzoic acid, perchloric acid, perborates, peracetates, persulfates, peroxides, or oxygen species and their free radicals, such as OH, 5 OOH, superoxide ( $O_2^-$ ) radicals, siglet oxygen, ozonide, dioxygenyl cation ( $O_2^+$ ), dioxiranes, dioxitanes, or Fremy radicals.

22. The multicomponent system of claim 1 or 2, characterized in that as component d), it contains aliphatic ethers and/or aryl-substituted alcohols, such as:

2,3- dimethoxybenzyl alcohol, 3,4-dimethoxybenzyl alcohol, 2,4- dimethoxybenzyl alcohol, 2,6-dimethoxybenzyl alcohol, homovanillyl alcohol, ethylene glycol monophenyl ether, 2- 5 hydroxybenzyl alcohol, 4-hydroxybenzyl alcohol, 4-hydroxy-3- methoxybenzyl alcohol, 2-

methoxybenzyl alcohol, 2,5- dimethoxybenzyl alcohol, 2,4-dimethoxybenzylamine, 2,4-dimethoxybenzylamine hydrochloride, veratryl alcohol, and coniferyl alcohol.

23. The multicomponent system of claim 1 or 2, characterized in that as component (d), it contains olefins (alkenes), such as:

2-allylphenol, 2-allyl-6-methylphenol, allylbenzene, 3,4-dimethoxypropanylbenzene, p-methoxystyrene, 1-allylimidazol, 1-vinylimidazol, styrene, stilbene, allylphenyl ether, cinnamic acid benzyl ester, cinnamic acid methyl ester, 2,4,6-triallyloxy-1,3,5-triazine, 1,2,4- trivinylcyclohexane, 4-allyl-1,2-dimethoxybenzene, 4-tert-butylbenzoic acid vinyl ester, squalene, benzoin allyl ether, cyclohexene, dihydropyran, and N-benzylcinnamic acid anilide.

24. The multicomponent system of claim 1 or 2, characterized in that as component (d), it contains phenol ethers, such as:

2,3-dimethoxybenzyl alcohol, 3,4- dimethoxybenzyl alcohol, 2,4-dimethoxybenzyl alcohol, 2,6- dimethoxybenzyl alcohol, homovanillyl alcohol, 4- hydroxybenzyl alcohol, 4-hydroxy-3-methoxybenzyl alcohol, 2- methoxybenzyl alcohol, 2,5-dimethoxybenzyl alcohol, 2,4-dimethoxybenzylamine, 2,4-dimethoxybenzylamine hydrochloride, veratryl alcohol, coniferyl alcohol, veratrol, and anisol.

25. The multicomponent system of of claim 1 or 2, characterized in that as component (d), it contains carbonyl compounds, such as:

4-aminobenzophenone, 4-acetylbiphenyl, benzophenone, benzil, benzophenone hydrazone, 3,4- dimethoxybenzaldehyde, 3,4-dimethoxybenzoic acid, 3,4- dimethoxybenzophenone, 4-dimethylaminobenzaldehyde, 4- acetylbiphenylhydrazone, benzophenone-4-carboxylic acid, benzoyl acetone, bis-(4,4-dimethylamino)benzophenone, benzoin, benzoin oxime,

- N-benzoyl-N-phenylhydroxylamine, 2- amino-5-chlorobenzophenone, 3-hydroxy-4-methoxybenzaldehyde, 4-methoxybenzaldehyde, anthraquinone-2-sulfonic acid, 4-methylaminobenzaldehyde, benzaldehyde, benzophenone-2- carboxylic acid, 3,3,'4,4'-
- 10 benzophenonetetracarboxylic acid dianhydride, (S)-(-)-2-(N-benzylpropyl)aminobenzo-henone, benzylphenyl acetic acid anilide, N-benzylbenzanilide, 4,4'- bis-(dimethylamino)-thiobenzophenone, 4,4-bis(diacetylamino)benzophenone, 2-chlorobenzophenone, 4,4'- dihydroxybenzophenone, 2,4-dihydroxybenzophenone, 3,5- dimethoxy-4-hydroxybenzaldehyde hydrazine, 4-hydroxybenzophenone, 4-methoxybenzophenone, 3,4- dihydroxybenzophenone, p-anisic acid, p-
- 15 anisic aldehyde, 3,4- dihydroxybenzaldehyde, 3,4-dihydroxybenzoic acid, 3,5- dimethoxy-4-hydroxybenzaldehyde, 3,5-dimethoxy-4- hydroxybenzoic acid, 4-hydroxybenzaldehyde, salicylaldehyde, vanillin, and vannilic acid.

26. The multicomponent system of claims 1 or 2, characterized in that as component (e), it contains benzotriazole as a free amine, in the case of the in situ generation or reaction mediation in cascade form for hydroxybenzotriazol.

27. The multicomponent system of claims 1 and 2, characterized in that as oxidoreductases, it contains enzymes originating in white rotting fungus [Phanerochaete chrysosporium], Trametes versicolor other fungi, bacteria, animals or plants, which enzymes are obtained from natural organisms or organisms that have been altered by gene technology.

28. The multicomponent system of claims 1 and 2, characterized in that as catalysts it contains modified enzymes, enzyme components, prosthetic groups or mimic substances, preferably heme groups or compounds containing heme groups.

29. The multicomponent system of claim 21, characterized in that as the oxidizing agents it contains oxygen, which is generated in situ by means of  $\text{H}_2\text{O}_2$ + catalase or other systems or  $\text{H}_2\text{O}_2$  from GOD+ glucose or other systems.

30. The multicomponent system of claims 1 to 29, characterized in that it contains cation-forming metal salts.

31. The multicomponent system of claim 30, characterized in that the cations are  $\text{Fe}^{2+}$ ,  $\text{Fe}^{3+}$ ,  $\text{Mn}^{2+}$ ,  $\text{Mn}^{3+}$ ,  $\text{Mn}^{4+}$ ,  $\text{Cu}^+$ ,  $\text{Cu}^{2+}$ ,  $\text{Ti}^{3+}$ ,  $\text{Ce}^{4+}$ ,  $\text{Mg}^{2+}$ , and  $\text{Al}^{3+}$ .

32. The multicomponent system of claims 1 and 31, characterized in that in addition it contains polysaccharides and/or proteins.

33. The multicomponent system of claims 1 to 32, characterized in that as polysaccharides it contains glucanes, mannanes, dextrans, levans, pectins, alginates, or vegetable rubbers, and/or its own polysaccharides formed by the fungi or produced in a mixed culture with yeasts, and as proteins, it contains gelatin or albumin.

34. The multicomponent system of claims 1 to 33, characterized in that as additives it contains simple sugar, oligomer sugar, amino acids, polyethylene glycols, polyethylene oxides, polyethylene imines and polydimethyl siloxanes.

35. A detergent containing the multicomponent system of one of claims 1 to 34.

36. The use of the multicomponent system of one of claims 1 to 35 as an additive to detergent formulations with detergent substances or detergent additives known per se.

37. The use of the multicomponent system of one of claims 1 to 36, characterized in that it is used at a pH value between 2 and 12, preferably between 4 and 10, and at a temperature between  $10^\circ\text{C}$  and  $60^\circ\text{C}$ , and preferably between  $20^\circ\text{C}$  and  $40^\circ\text{C}$ .



Docket No.  
CRZ-96631

# Declaration and Power of Attorney For Patent Application

## English Language Declaration

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name,

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled

**MULTICOMPONENT SYSTEMS FOR USE WITH DETERGENT SUBSTANCES**

the specification of which

(check one)

☐ is attached hereto.

☒ was filed on June 19, 1996 as United States Application No. or PCT International Application Number PCT/DE96/02658 and was amended on \_\_\_\_\_

(if applicable)

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose to the United States Patent and Trademark Office all information known to me to be material to patentability as defined in Title 37, Code of Federal Regulations, Section 1.56.

I hereby claim foreign priority benefits under Title 35, United States Code, Section 119(a)-(d) or Section 365(b) of any foreign application(s) for patent or inventor's certificate, or Section 365(a) of any PCT International application which designated at least one country other than the United States, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate or PCT International application having a filing date before that of the application on which priority is claimed.

Prior Foreign Application(s)

Priority Not Claimed

_____ (Number)	_____ (Country)	_____ (Day/Month/Year Filed)	<input type="checkbox"/>
_____ (Number)	_____ (Country)	_____ (Day/Month/Year Filed)	<input type="checkbox"/>
_____ (Number)	_____ (Country)	_____ (Day/Month/Year Filed)	<input type="checkbox"/>

008020" T0462060

I hereby claim the benefit under 35 U.S.C. Section 119(e) of any United States provisional application(s) listed below:

_____ (Application Serial No.)	_____ (Filing Date)
_____ (Application Serial No.)	_____ (Filing Date)
_____ (Application Serial No.)	_____ (Filing Date)

I hereby claim the benefit under 35 U. S. C. Section 120 of any United States application(s), or Section 365(c) of any PCT International application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International application in the manner provided by the first paragraph of 35 U.S.C. Section 112, I acknowledge the duty to disclose to the United States Patent and Trademark Office all information known to me to be material to patentability as defined in Title 37, C. F. R., Section 1.56 which became available between the filing date of the prior application and the national or PCT International filing date of this application:

_____ (Application Serial No.)	_____ (Filing Date)	_____ (Status) (patented, pending, abandoned)
_____ (Application Serial No.)	_____ (Filing Date)	_____ (Status) (patented, pending, abandoned)
_____ (Application Serial No.)	_____ (Filing Date)	_____ (Status) (patented, pending, abandoned)

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

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POWER OF ATTORNEY: As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and transact all business in the Patent and Trademark Office connected therewith. (list name and registration number)

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Second inventor's signature
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